AFCI Fuel Development Update: Metallic Fuel & Irradiation Testing

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Outline

- Review of Issues
- Fabrication of Metal Fuel for AFC-1
- Characterization (results to date)
  - Pu-60Zr
  - Pu-40Zr
  - Pu-12Am-40Zr
- Irradiation Testing
- Summary

Fuel Issues Associated with Transmutation

- **Fuel performance**
  - High He generation rate during (and after) irradiation due to Am-241 and Cm-244
  - Am transport during irradiation
  - Actinide compatibility with cladding
  - Formation of low-melting-temperature phases
- **Fabrication and fuel recycle**
  - Compatibility with remote fuel recycle (incl process complexity concerns
  - Loss goal < 0.1% during refabrication
  - Volatility of Am during thermal processing
  - Cm is hard to handle, high α-decay heat

Inert Matrix (Zr) and Actinide (Pu,Np,Am) Single Element Phases

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$Im\bar{3}m$ (bcc)	ε-Pu 483°C	γ-Np 576°C	γ-Am 1077°C	β-Zr 863°C
$Fm\bar{3}m$ (fcc)	δ-Pu 320°C	-	β-Am 769°C	-
$P6_3/mmc$ (hcp)	-	-	α-Am	α-Zr
Fddd	γ-Pu	-	-	-
$P42_12$	-	β-Np	-	-
Pnma	-	α-Np	-	-

Fuel Fabrication Challenges

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Phase Relations in Metallic Systems

Pu-Zr	**documented**
Pu-Am	**documented in Pu rich region**
Pu-Np	**documented**
Np-Zr	**limited data (conflicting results)**
Np-Am	**limited data**
Am-Zr	**no data**
Pu-Np-Am	**limited theoretical**
Pu-Np-Zr	**no data**
Pu-Am-Zr	**no data**
Np-Am-Zr	**no data**
Pu-Np-Am-Zr	**no data**

Pu-Zr Based Metallic Alloys

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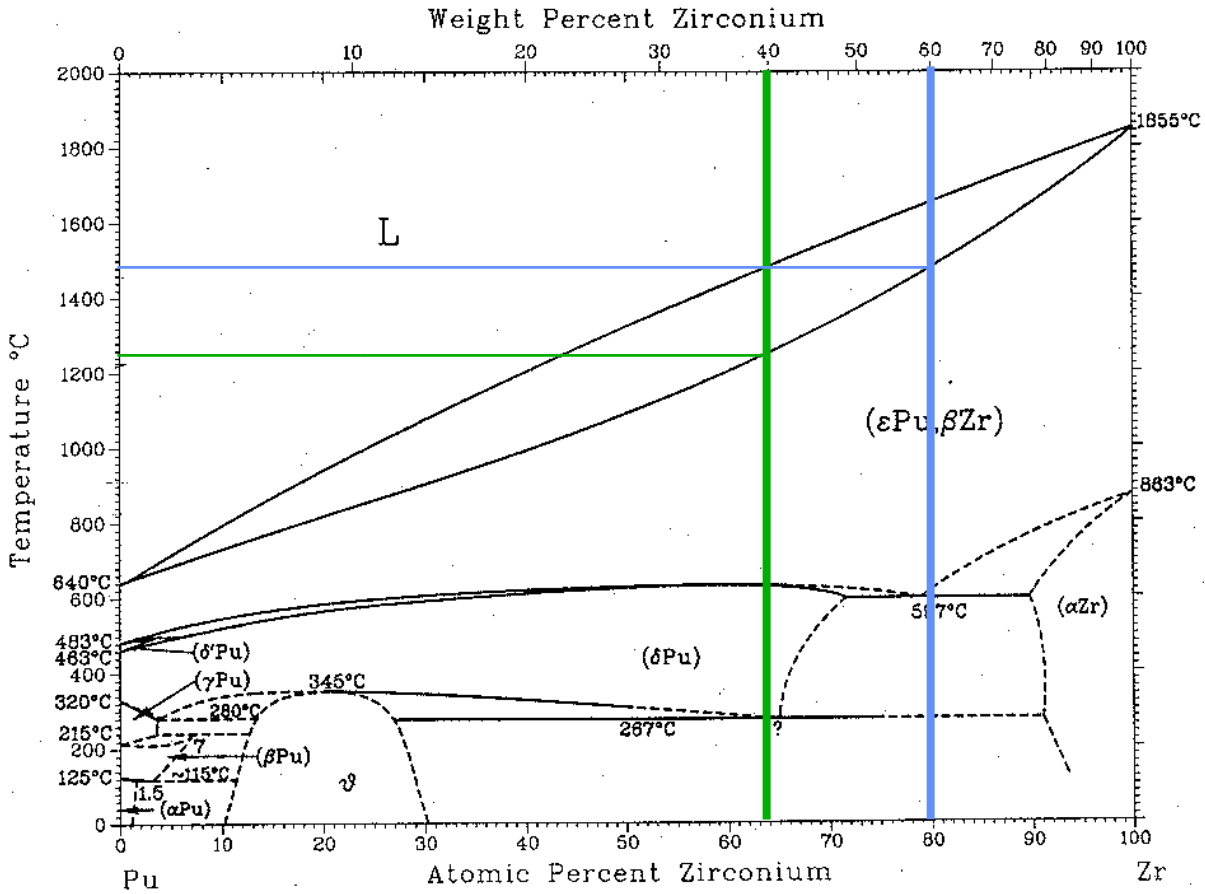
Pu-Zr based fuels for irradiation testing in AFC-1

- Pu-40Zr
- Pu-Am-40Zr
- Pu-Am-Np-40Zr
- Pu-Np-40Zr

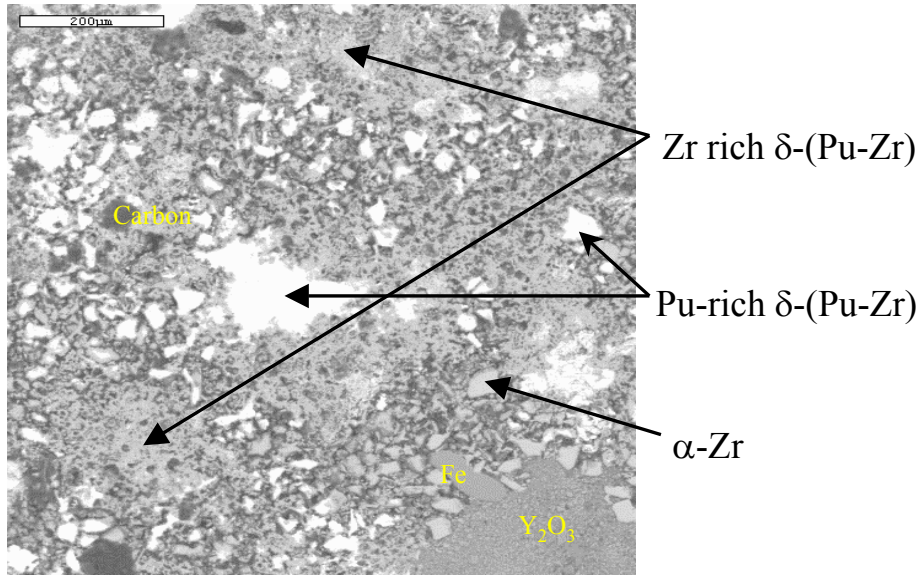
- Pu-60Zr

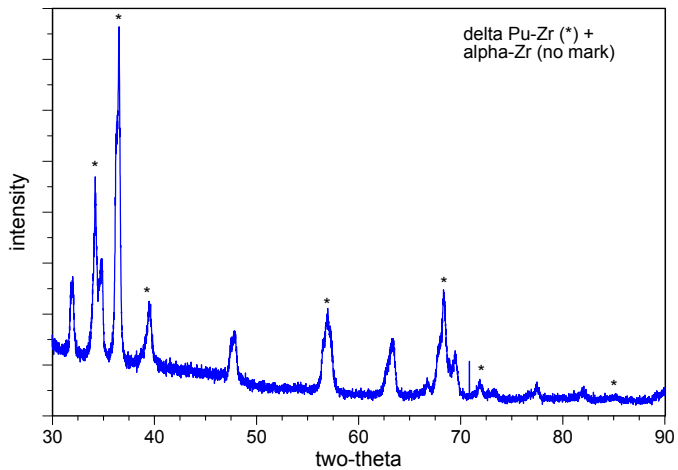


Binary Alloy Phase Diagrams 2nd ed. Vol. 3 Ed. T.B. Massalski, ASM Int'l (1990)

Powder Metallurgy Route to Pu-Zr

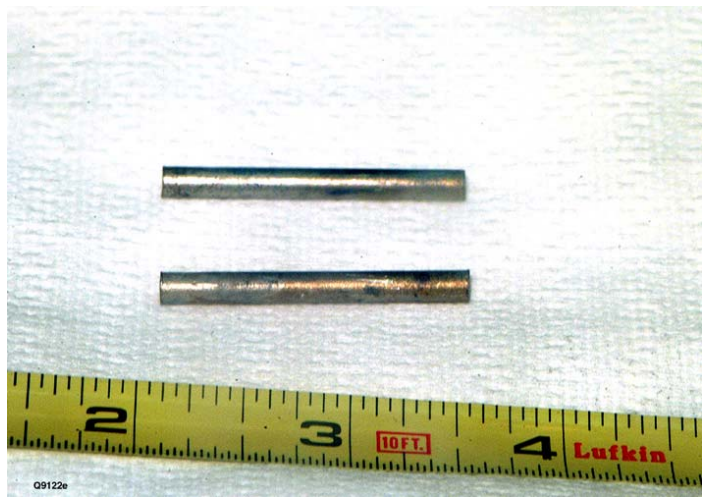
- **Developed as low temperature fabrication route to prevent Am loss**
  - **Hot pressing**
- **Inhomogeneous microstructure after processing at ~ 800°C**
- **High impurity content**
- **Complex process - impractical remote process scale-up**
  - **Powder handling**
  - **Material loss**
  - **Time**

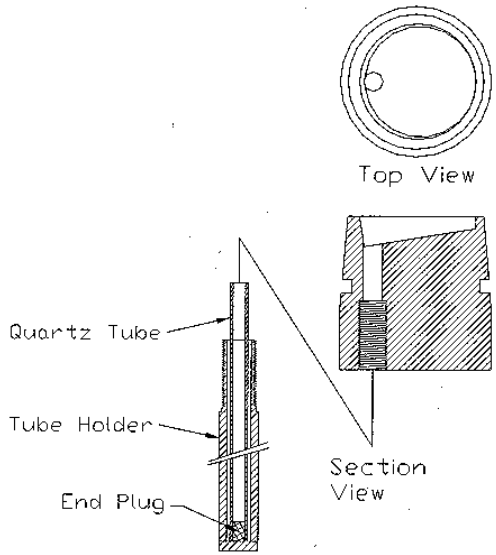




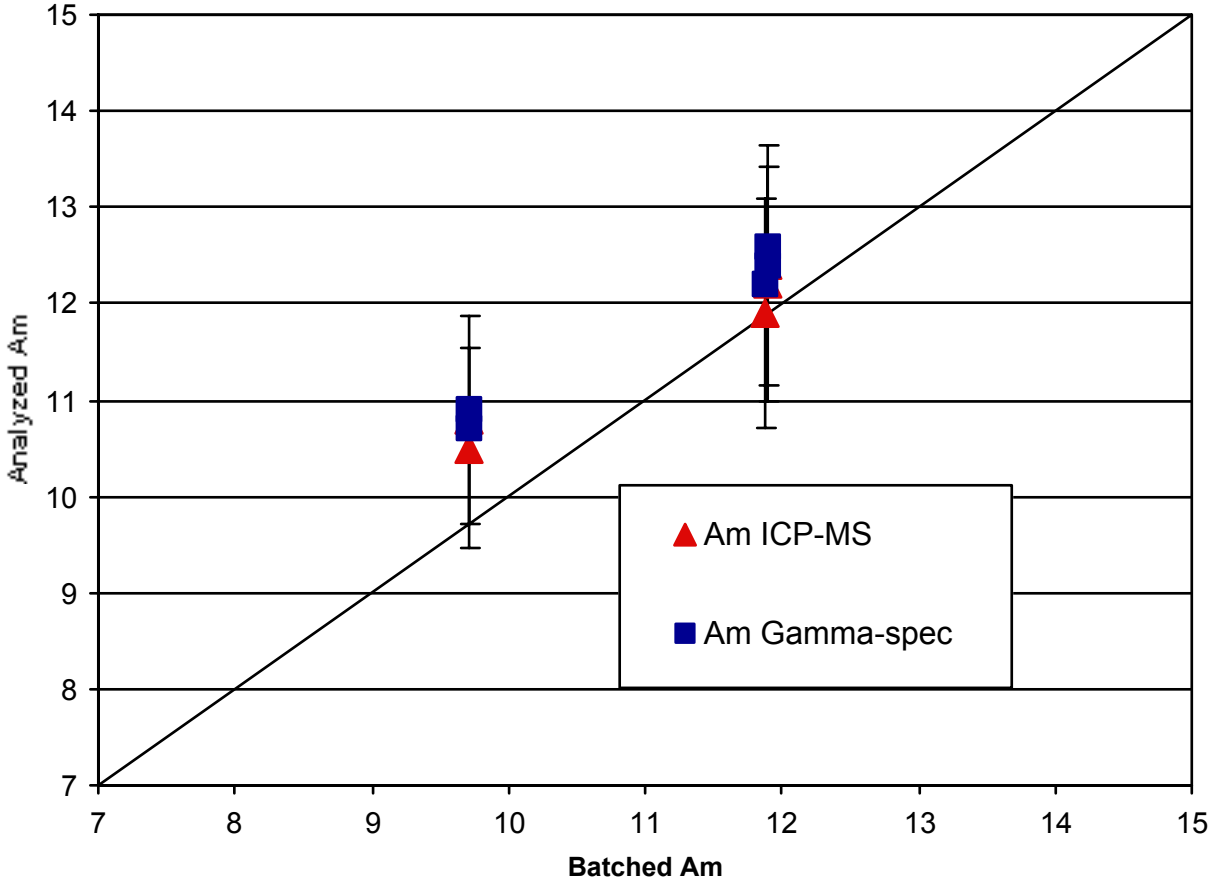
Arc Melting to Cast Fuel

- Simple process
- Short melt times at high temperature
  - Total melt times < 60 seconds
  - Temperature > 2000°C
- Good alloy homogeneity
- Low americium loss



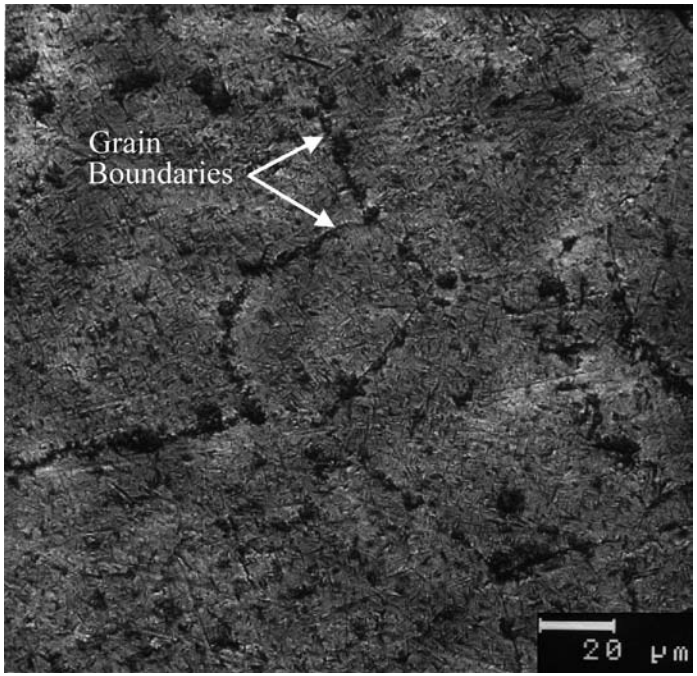


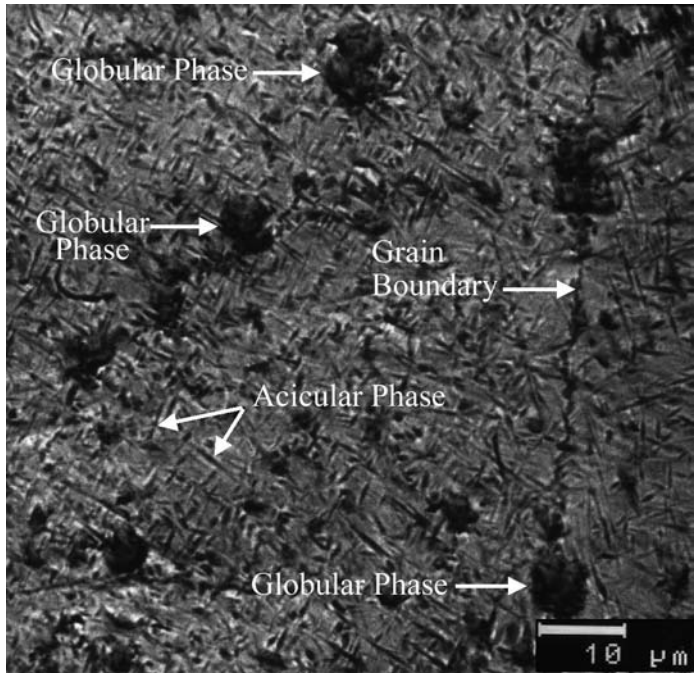
Americium Loss Data



- Results from arc-casting Pu-Am-Zr and Pu-Am-Np-Zr
- Other rapid melting techniques such as induction skull melting (ISM) can also have melt cycle times on the order of 60 seconds
- Am loss during fabrication is not a critical issue for metal fuel

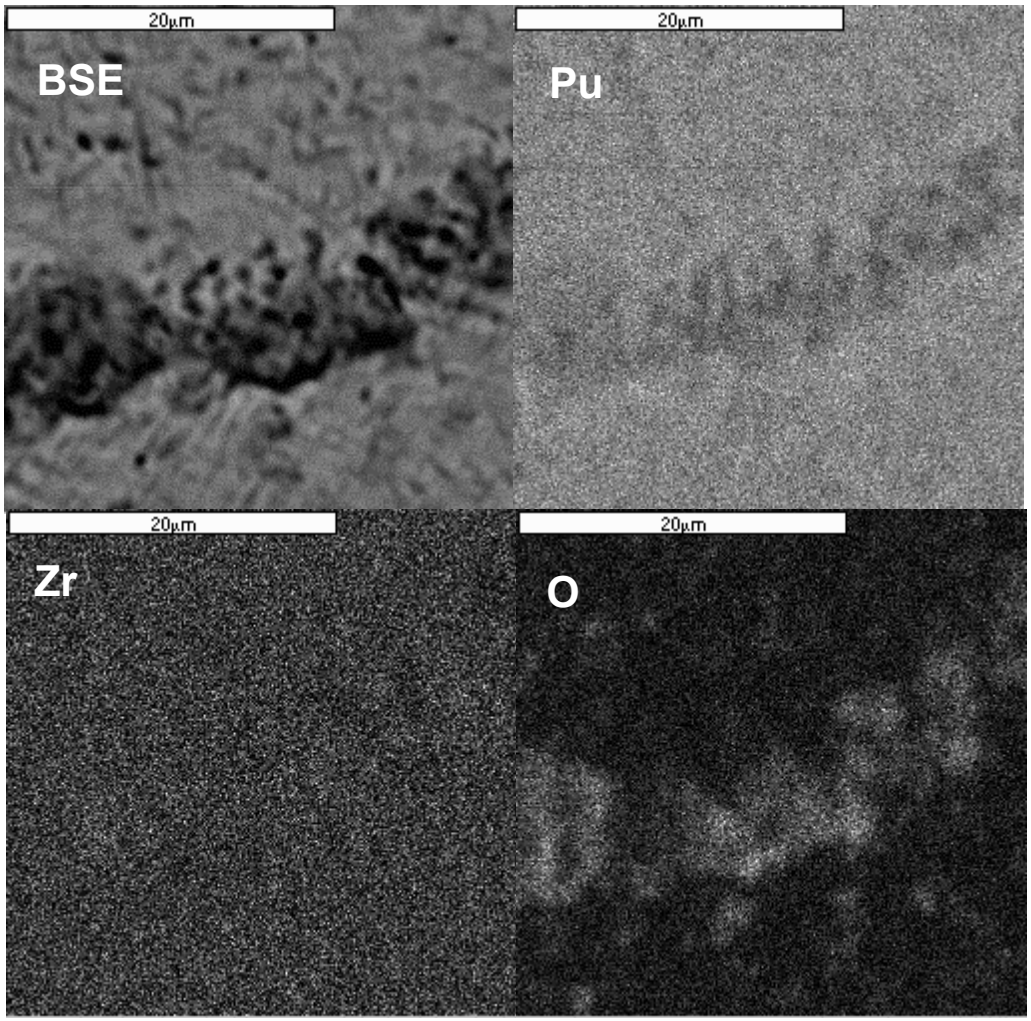
Pu-60Zr



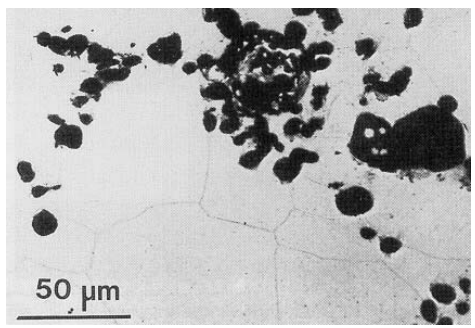


- **As-cast microstructure – lowest fuel fabrication cost**
- **SEM images show multi-phase structure**
  - **Matrix, acicular phase**
  - **Grain boundaries decorated with second phase(s)**
- **Measured density = 8.5 g/cm^3**

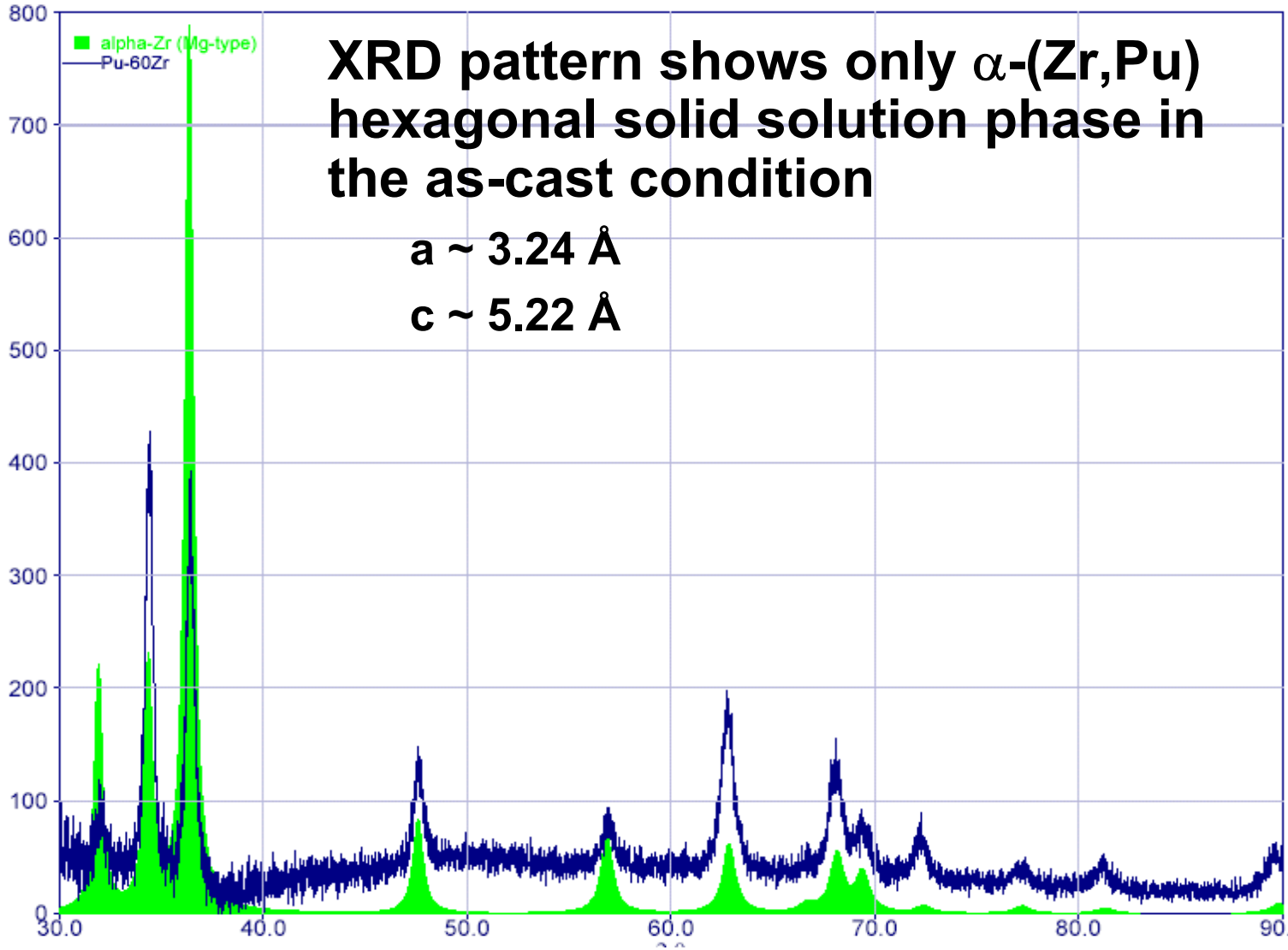
As-cast Pu-60Zr



- **Chemistry differences between acicular phase and matrix phase cannot be resolved using SEM/EDS**
- **EDS indicates that grain boundary phase is plutonium depleted and oxygen rich**
- **Uniform distribution of zirconium**
- **Similar phase in U-20Pu-10Zr**



Pu-60Zr XRD



XRD pattern shows only α-(Zr,Pu) hexagonal solid solution phase in the as-cast condition

- a ~ 3.24 Å
- c ~ 5.22 Å

Pu-60Zr Thermal Analysis

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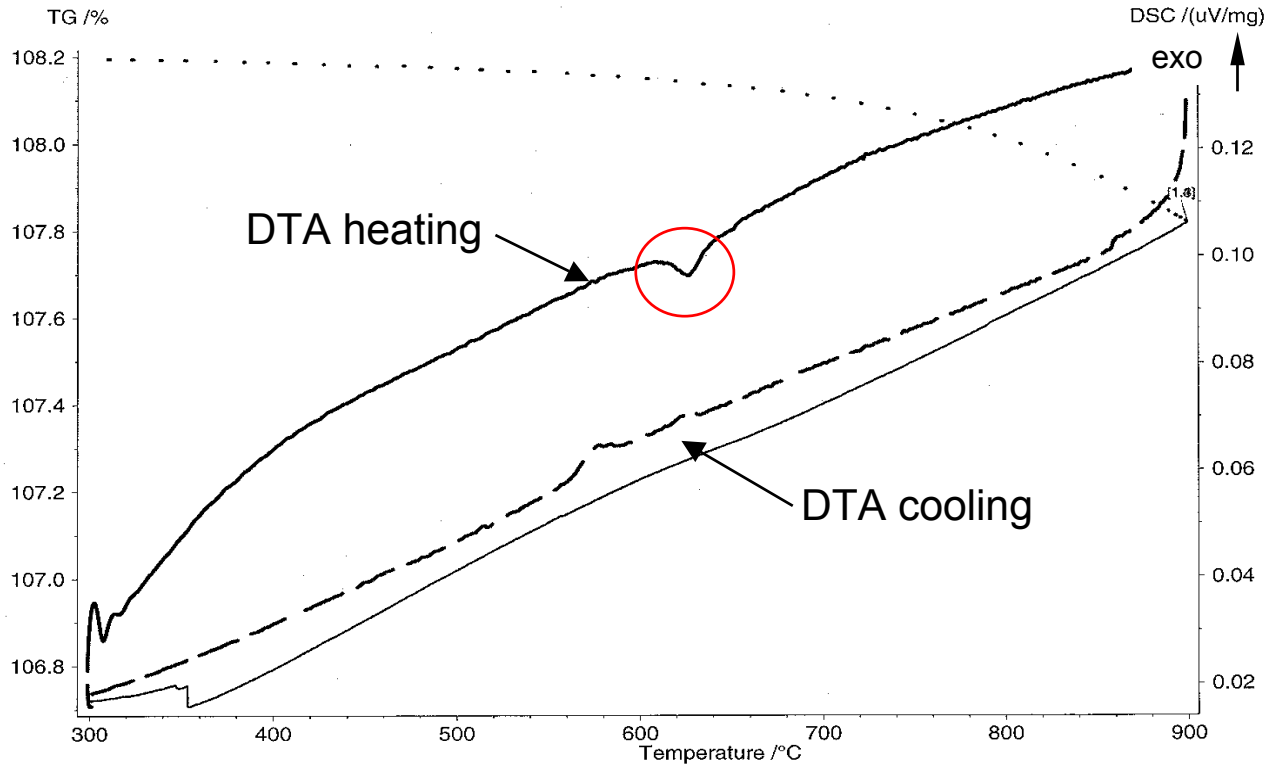
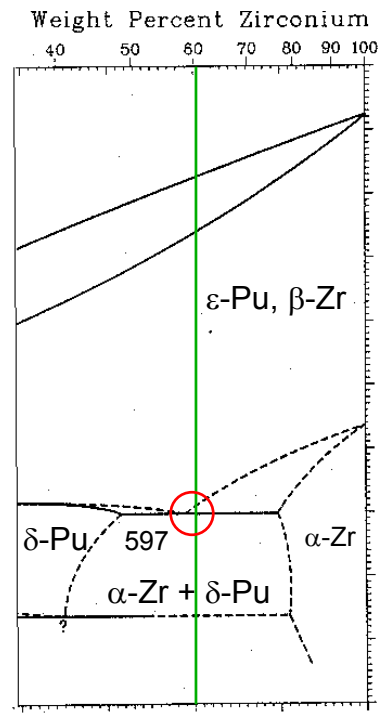


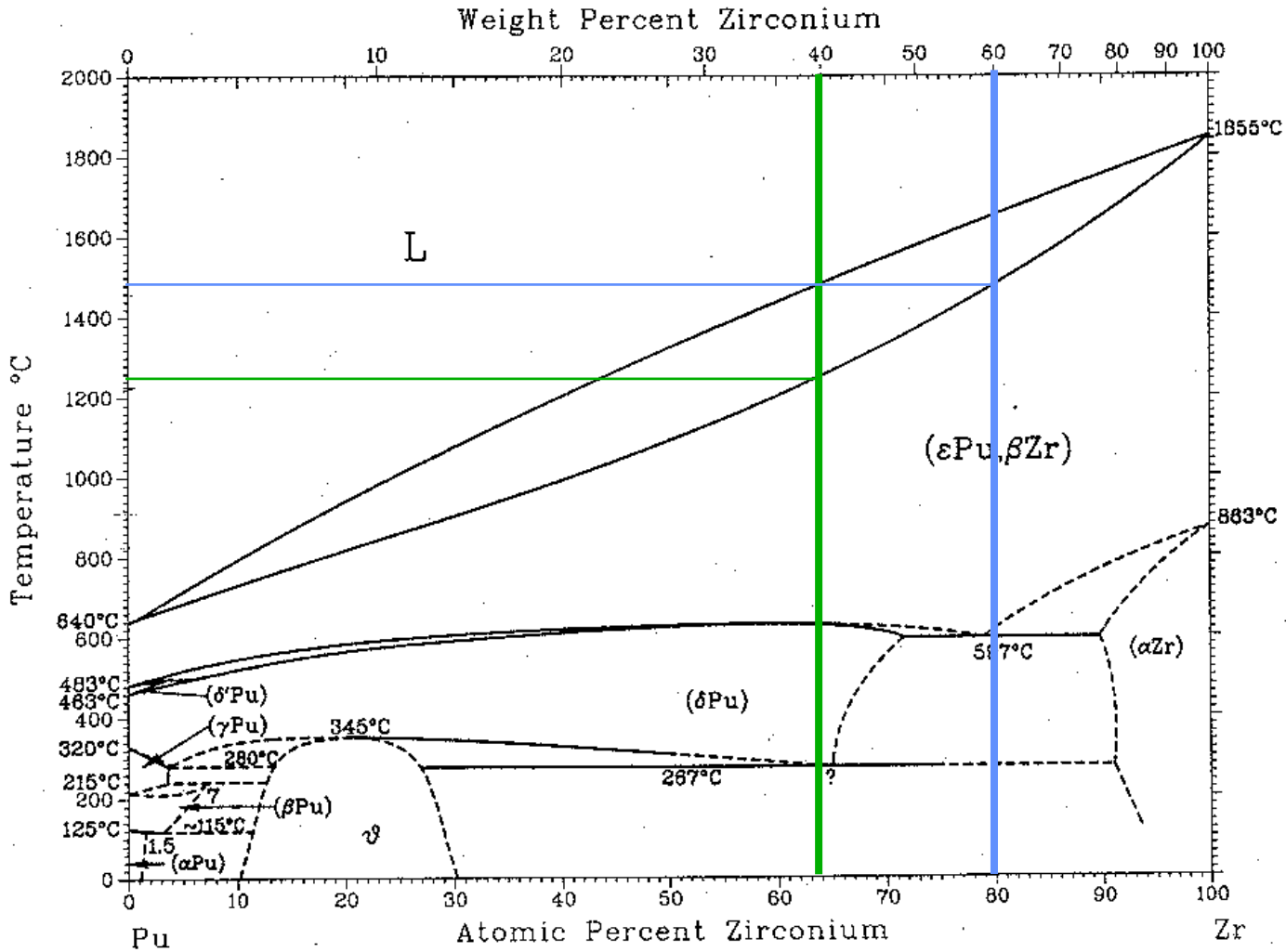


DTA shows repeatable transformation

α-Zr + δ-Pu $\longrightarrow$ ε-Pu, β-Zr solid solution

Oxidation prevents further interpretation

Pu-40Zr



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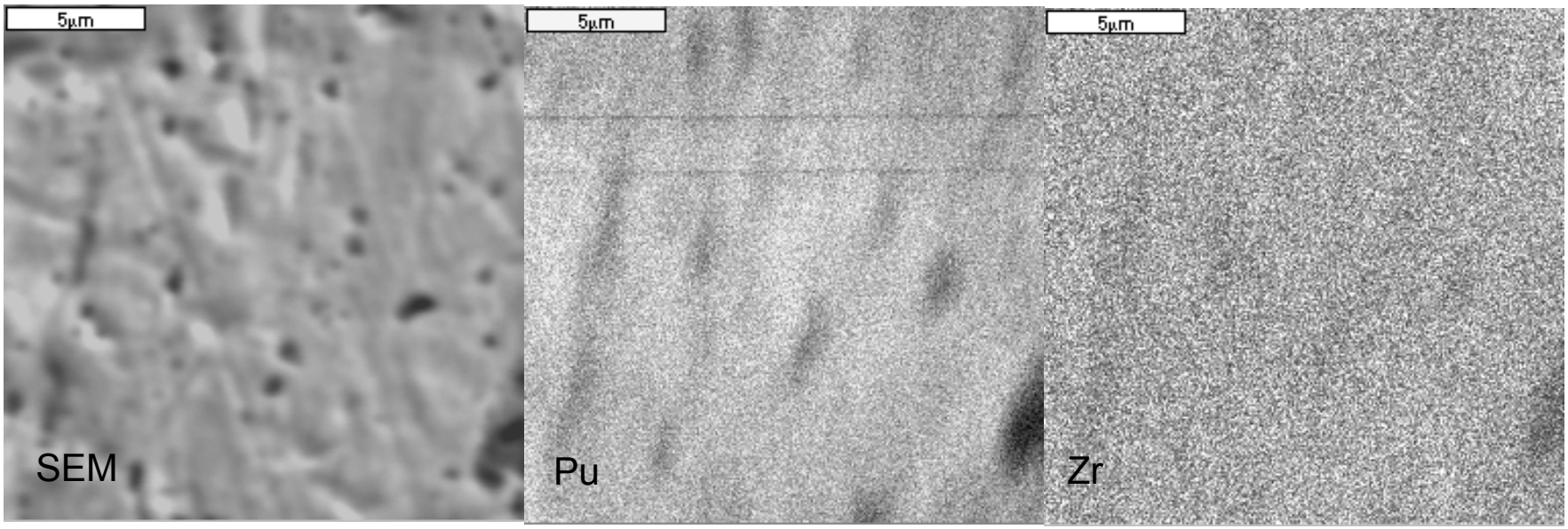
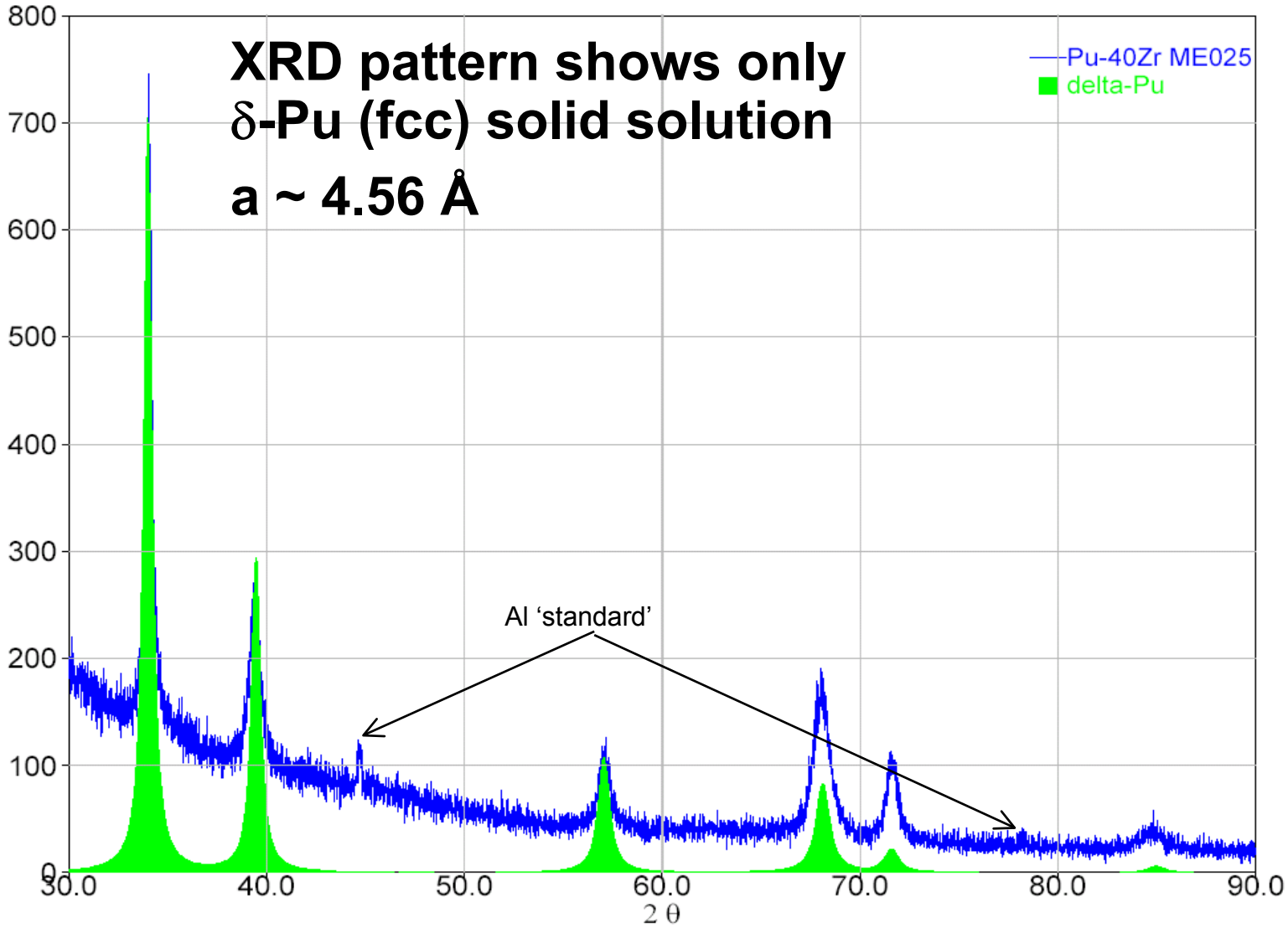
Pu-40Zr



- **Single phase alloy**
- **SEM/EDS shows uniform distribution of Pu-Zr**
- **Some porosity present in as-cast microstructure**
- **Measured density = 10.1 g/cm^3**

Pu-40Zr XRD


XRD pattern shows only
δ-Pu (fcc) solid solution
a ~ 4.56 Å
Pu-40Zr ME025
delta-Pu
Al 'standard'
800
700
600
500
400
300
200
100
0
30.0
40.0
50.0
60.0
70.0
80.0
90.0
2 θ

Pu-40Zr Thermal Analysis

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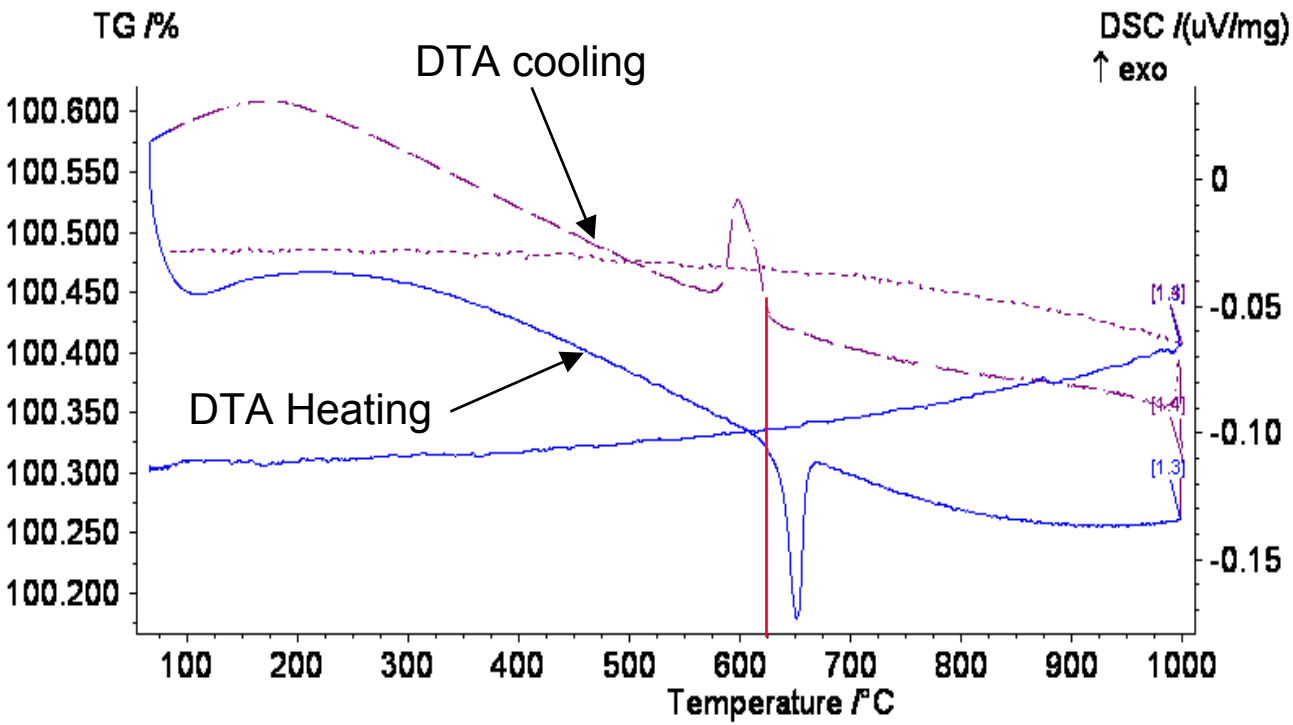

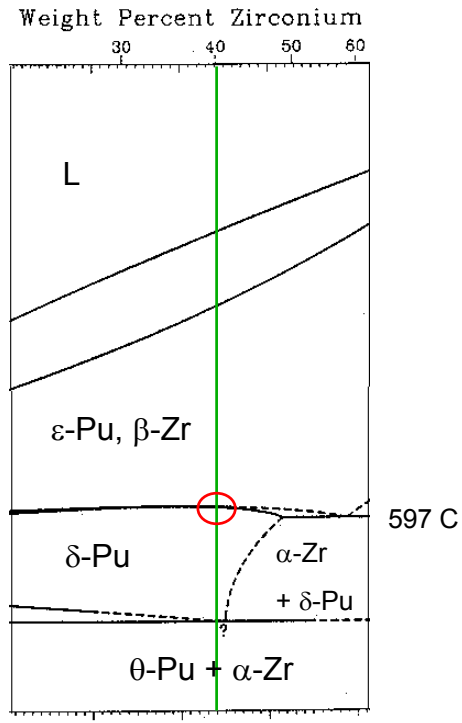

DTA shows repeatable transformation

δ-Pu (fcc) ⟶ ε-Pu, β-Zr (bcc) ~625°C

Pu-Am-Zr Phase Equilibria

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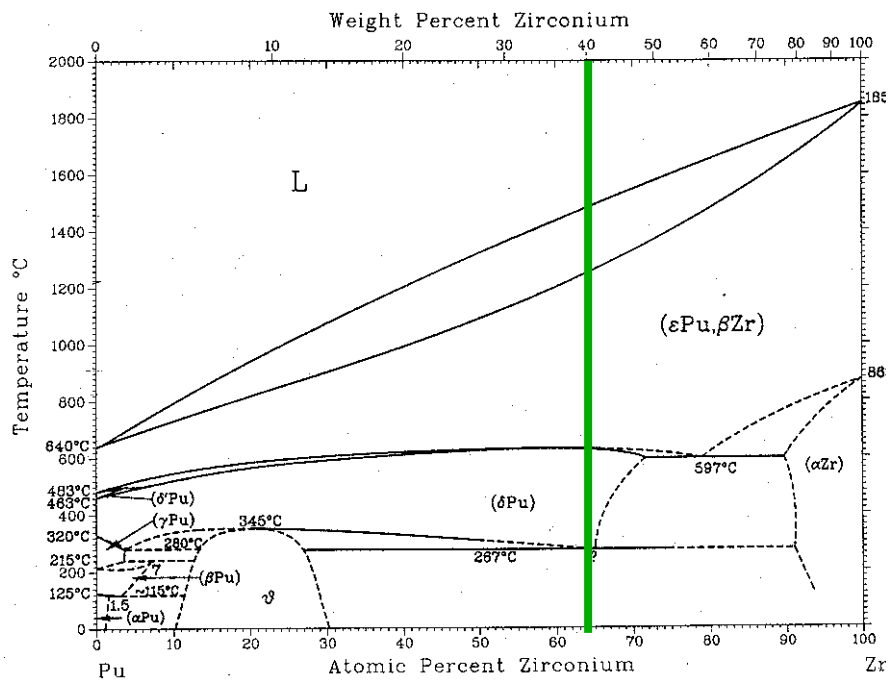
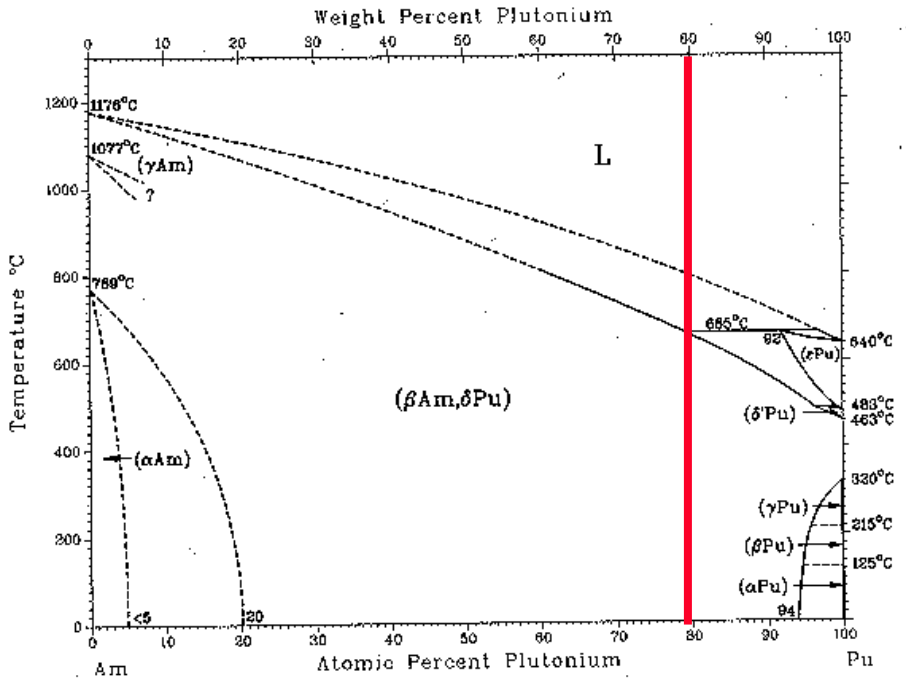
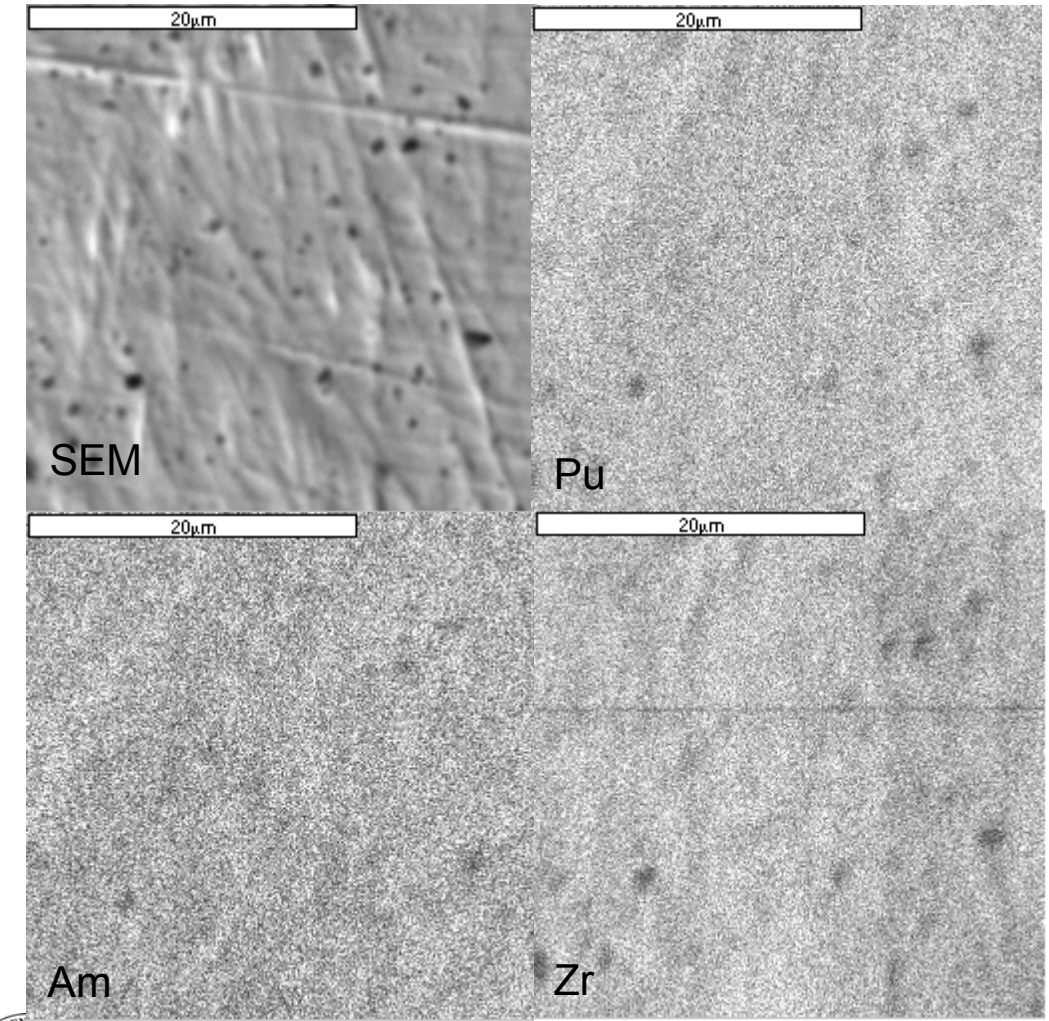




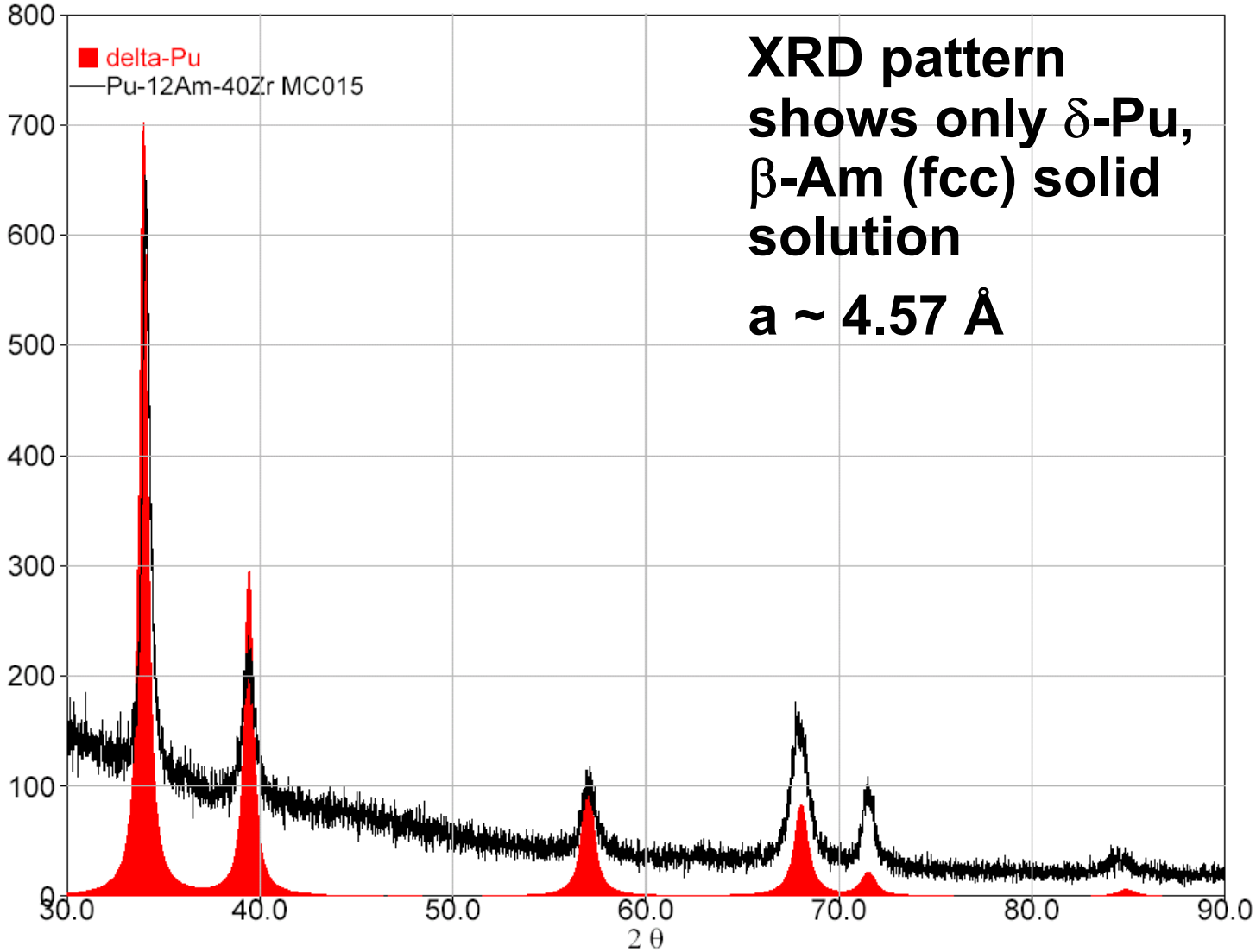
- **Wide region of δ-Pu, β-Am (fcc) stability**
- **Am increases binary alloy solidus temperature**

Pu-12Am-40Zr



- **SEM/EDS shows a single phase alloy with a uniform distribution of Pu, Am, and Zr**
- **Measured density = 9.9 g/cm^3**

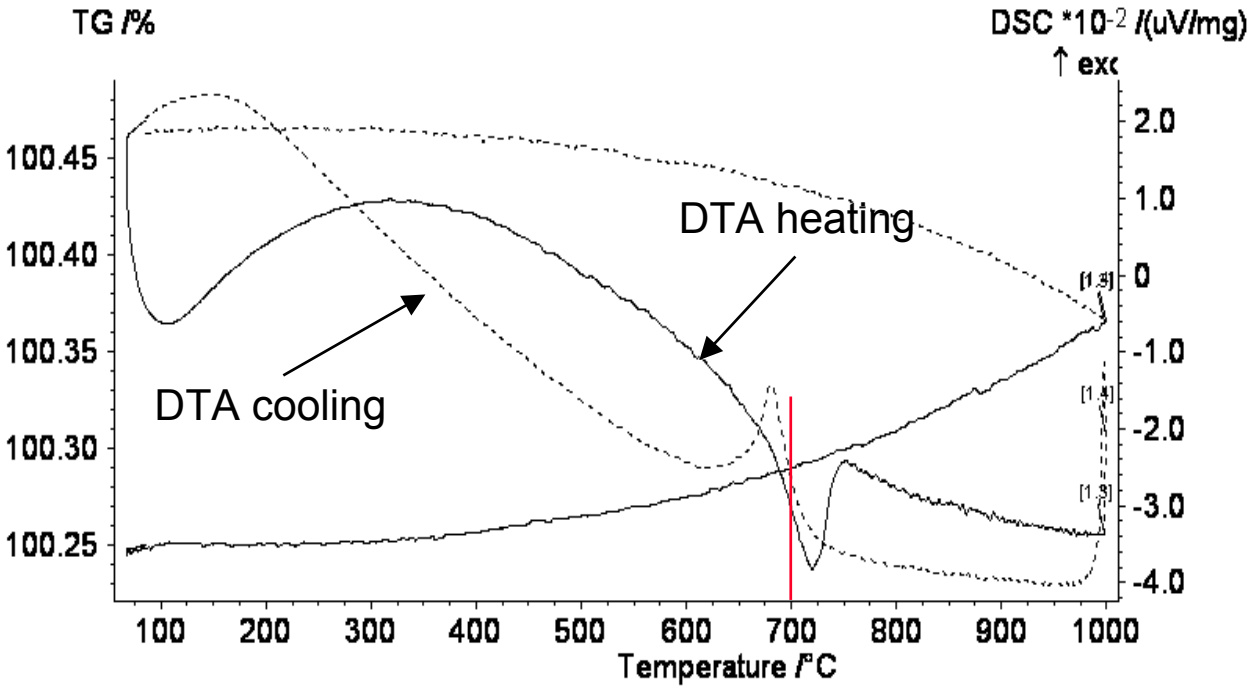
Pu-12Am-40Zr XRD

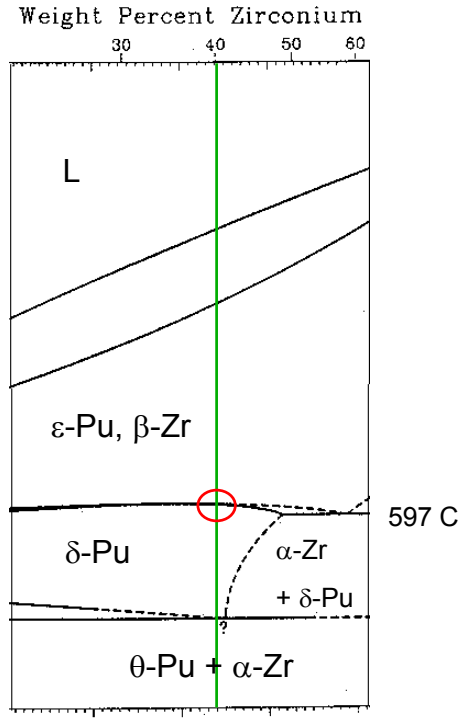


XRD pattern shows only δ-Pu, β-Am (fcc) solid solution

a ~ 4.57 Å

Pu-12Am-40Zr Thermal Analysis





- **Higher transition temperature (~700°C) relative to Pu-40Zr (~625°C) may indicate expansion of the δ-Pu phase field in Am-bearing alloy**

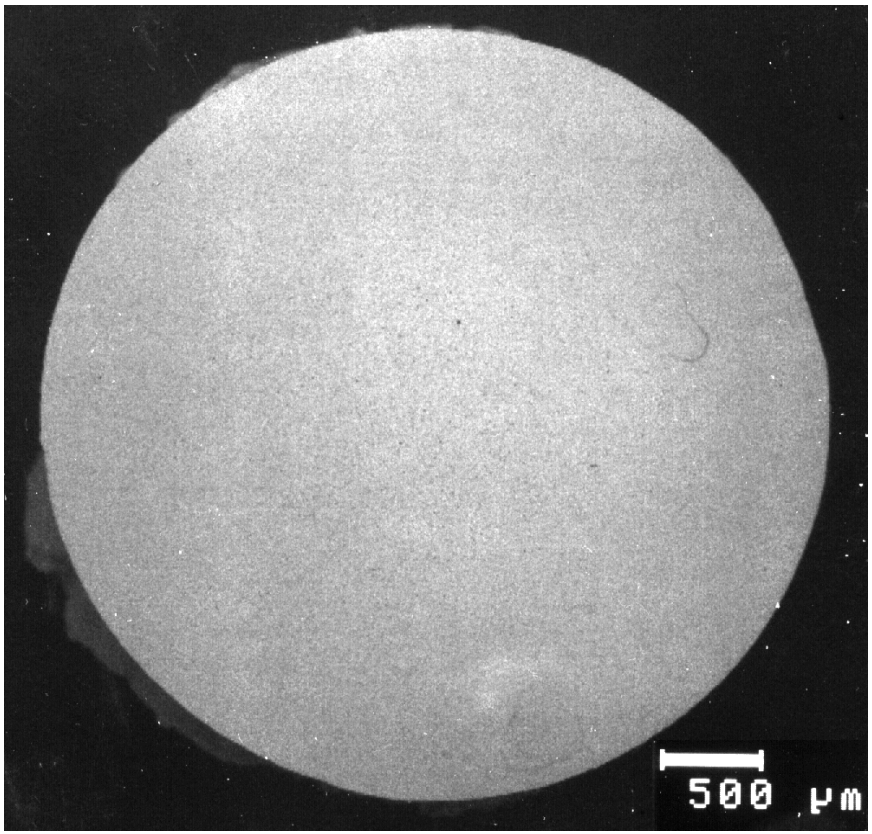
Processing, Chemical and Physical Characteristics for Pu-Np-Zr samples

Pin Number	Composition Wt. %			Number of Melts		Average Density g/cm3	Theoretical Density* g/cm3
	Pu	Np	Zr	Homo-genization	Casting		
MB008	49.8	8.8	40.4	5	2	10.33	10.82
MB010	50.3	7.9	41.6	7	3	10.27	10.72

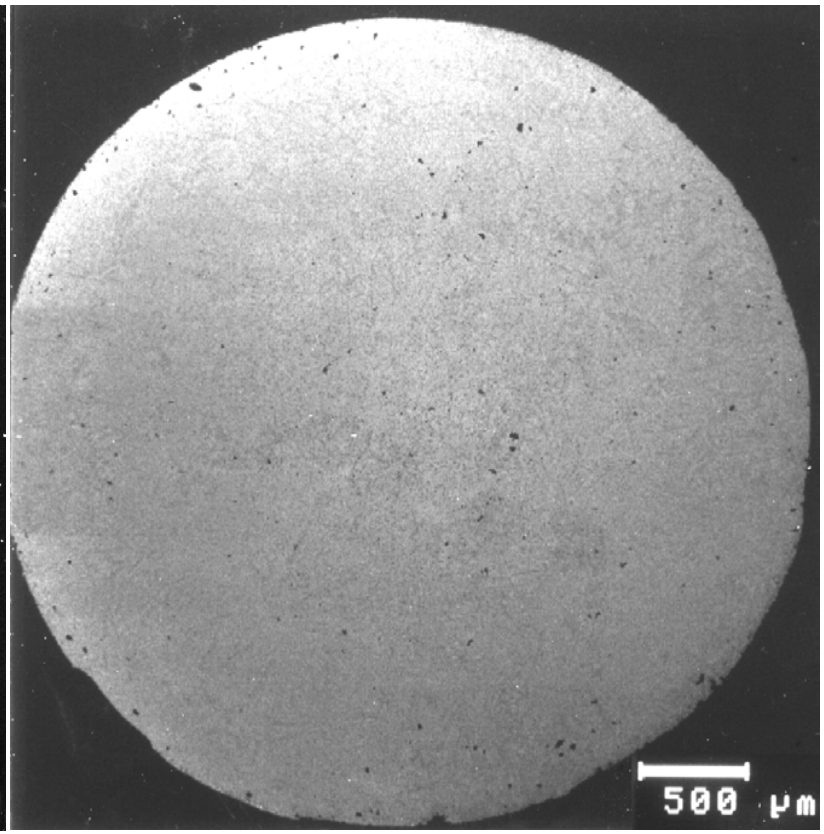
* each composition contains ~0.2 wt.% Am and assumes ideal solution behavior

Comparison SEM Study on Pu-40Zr to Pu-10Np-40Zr As-Cast Microstructures

Pu-40Zr



Pu-10Np-40Zr



Comparison SEM Study on Pu-40Zr to Pu-10Np-40Zr As-Cast Microstructures

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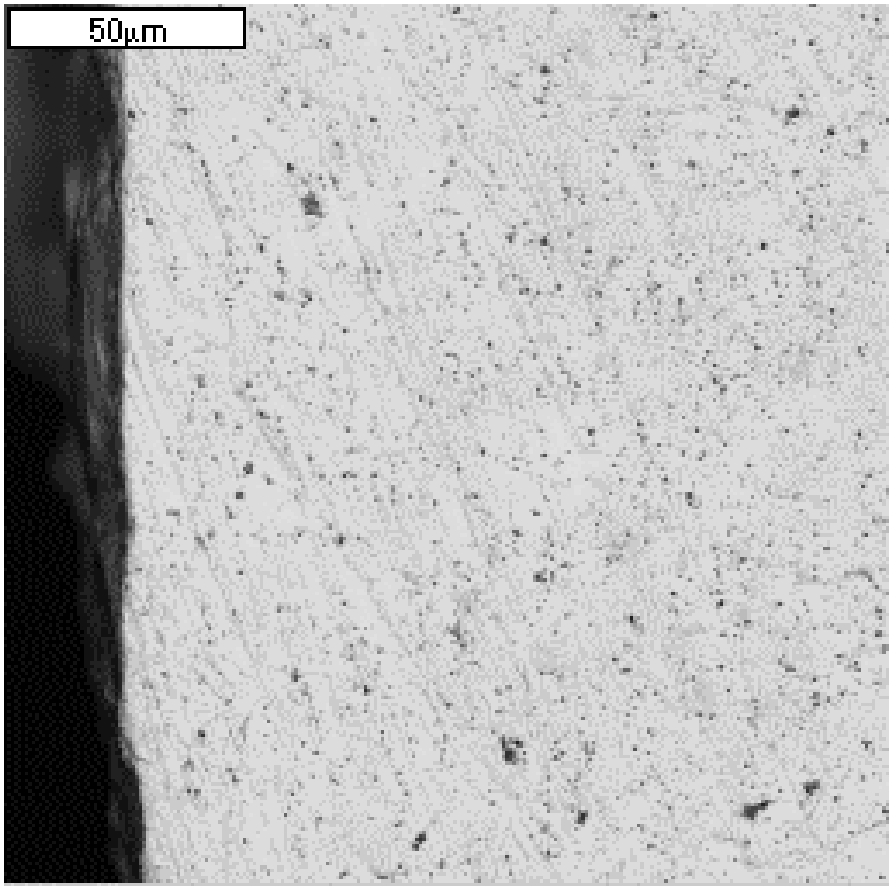
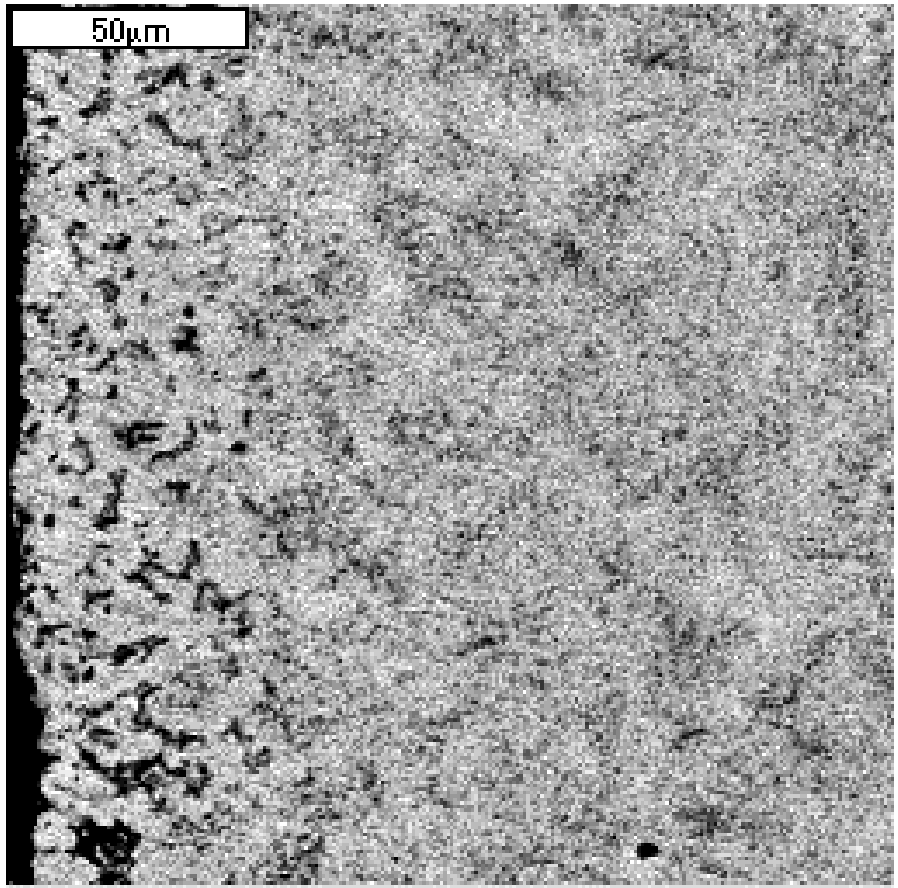
Pu-40Zr edge



Pu-10Np-40Zr edge



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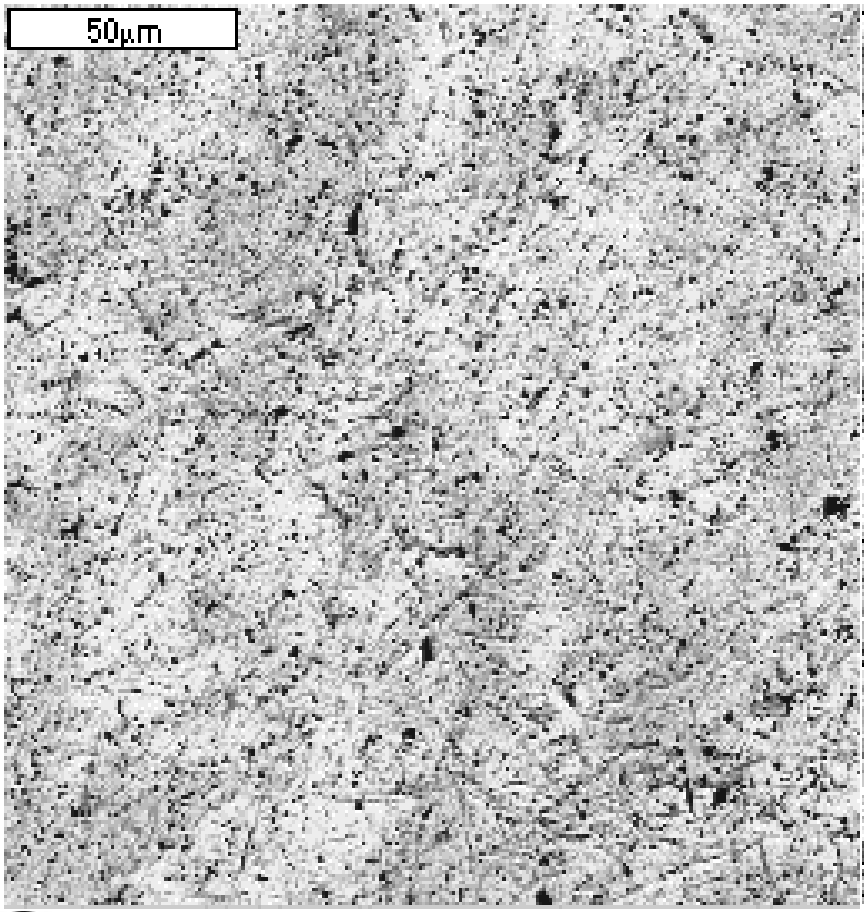
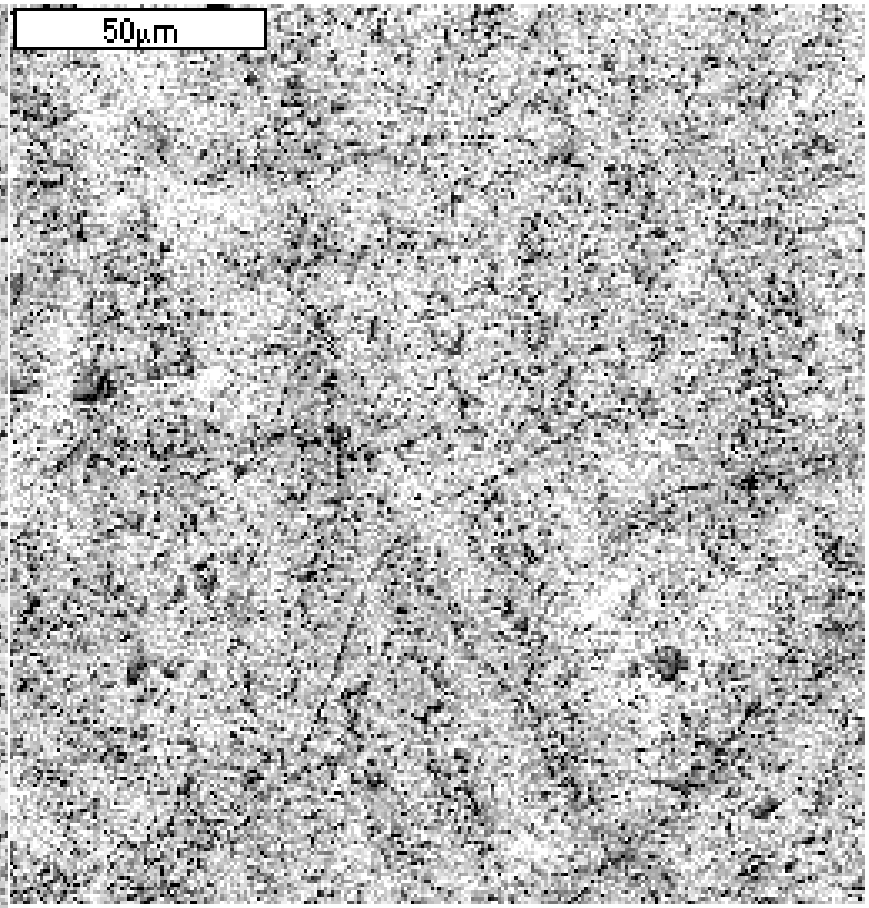
Pu-40Zr

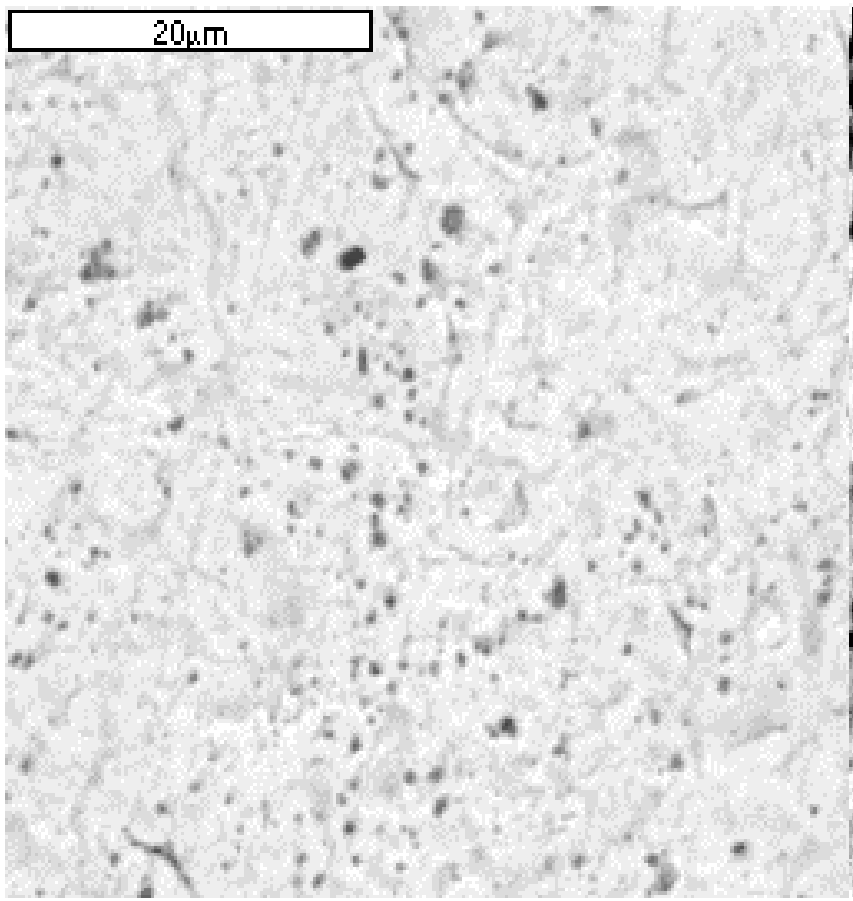


Pu-10Np-40Zr

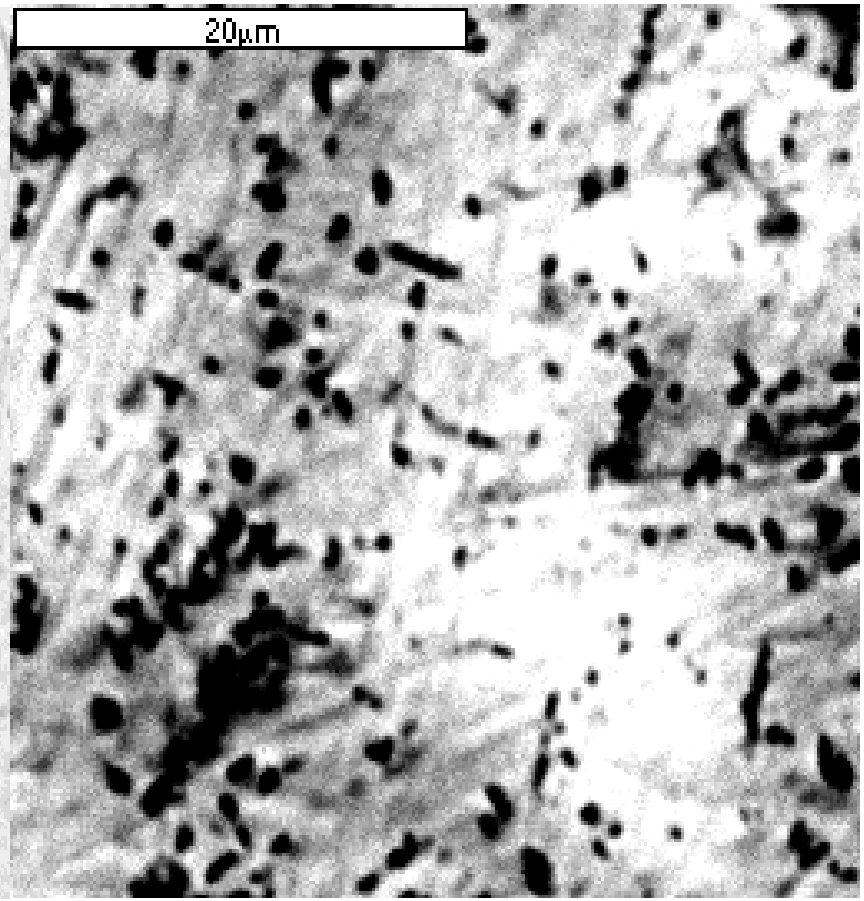


Comparison SEM Study on Pu-40Zr to Pu-10Np-40Zr As-Cast Microstructures

Pu-40Zr

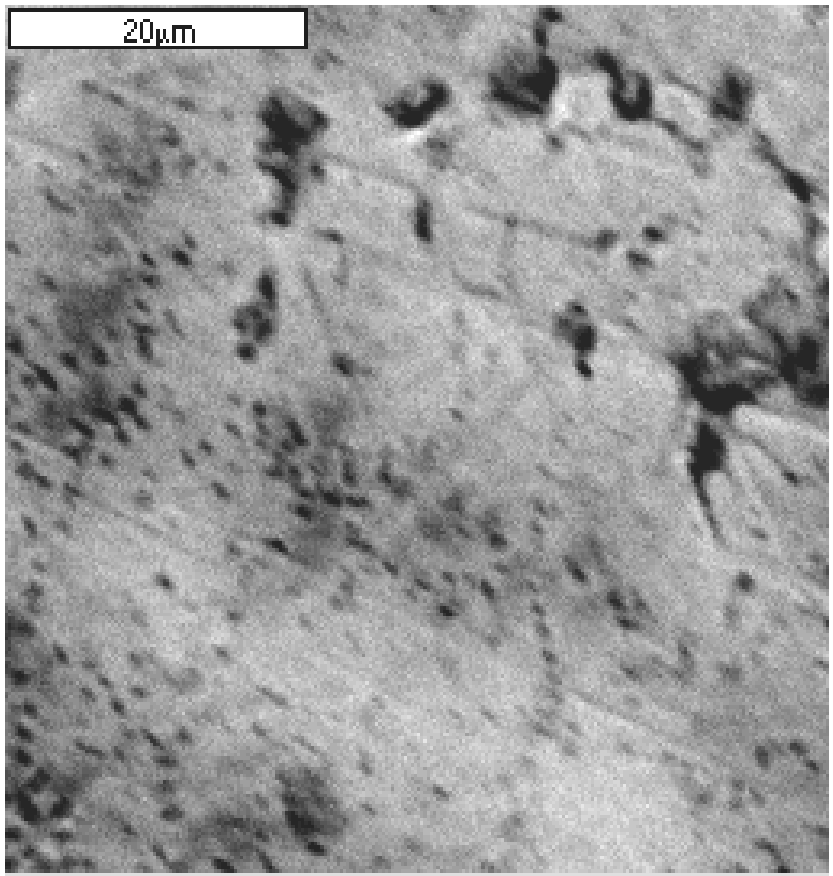


Pu-10Np-40Zr

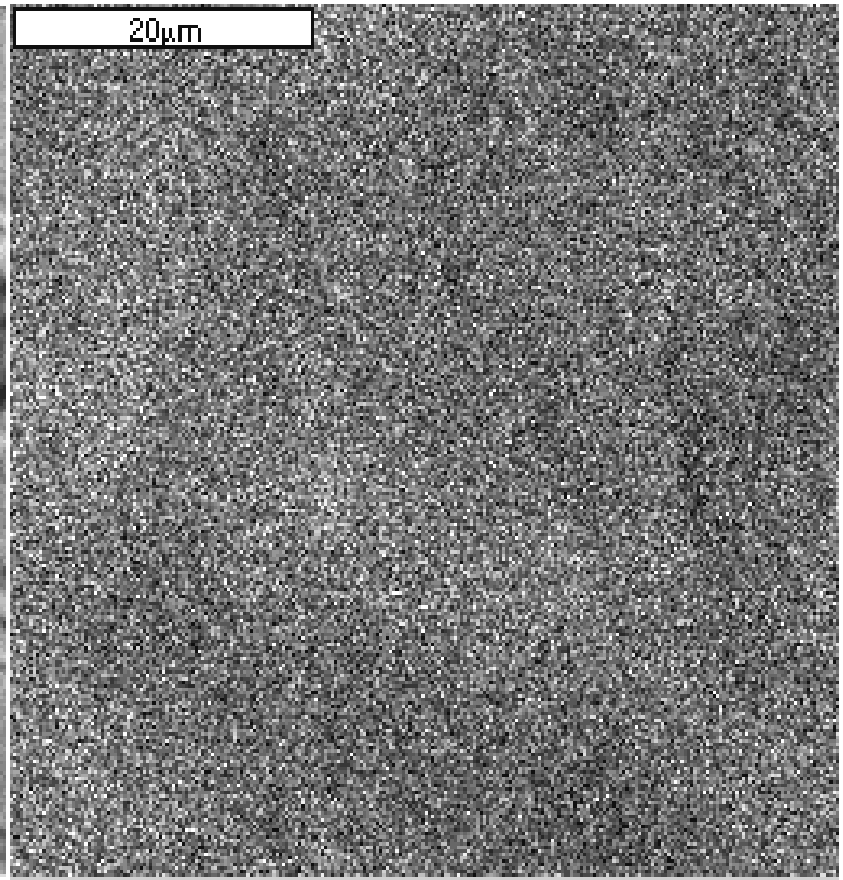


X-ray Maps of As-Cast Pu-10Np-40Zr

Backscatter Image



Zr



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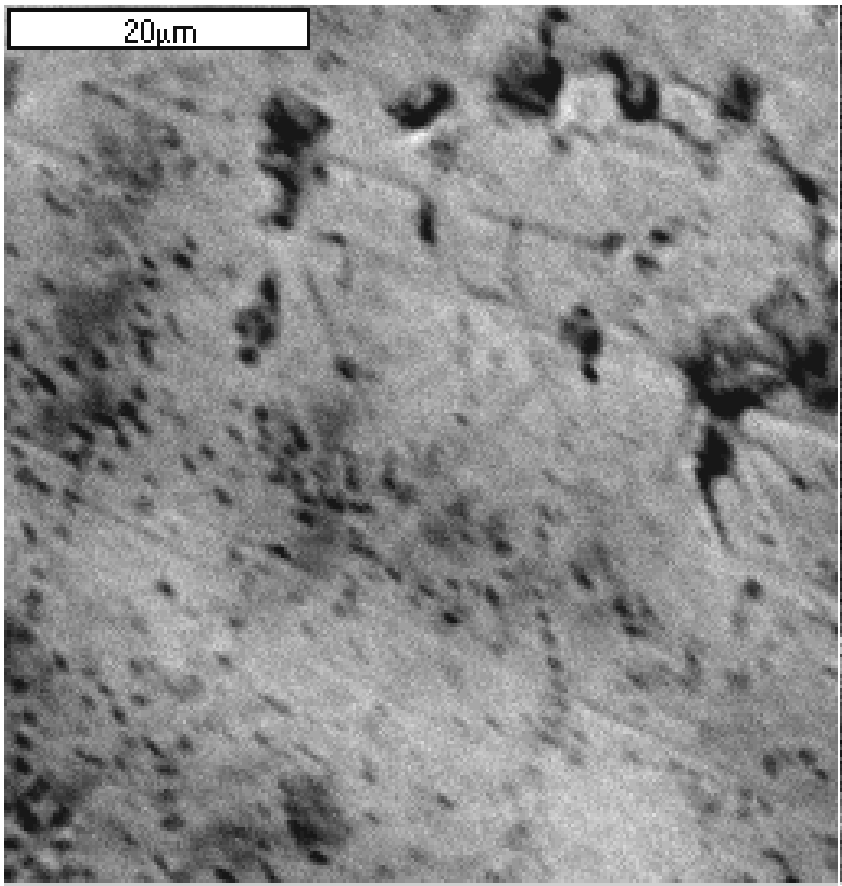
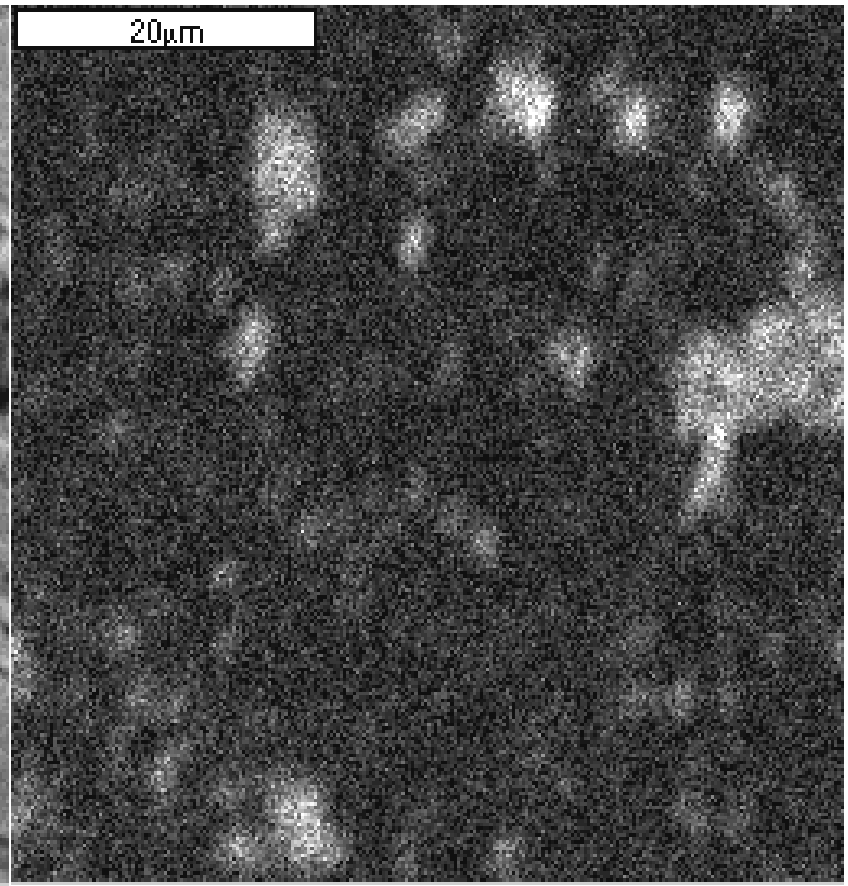
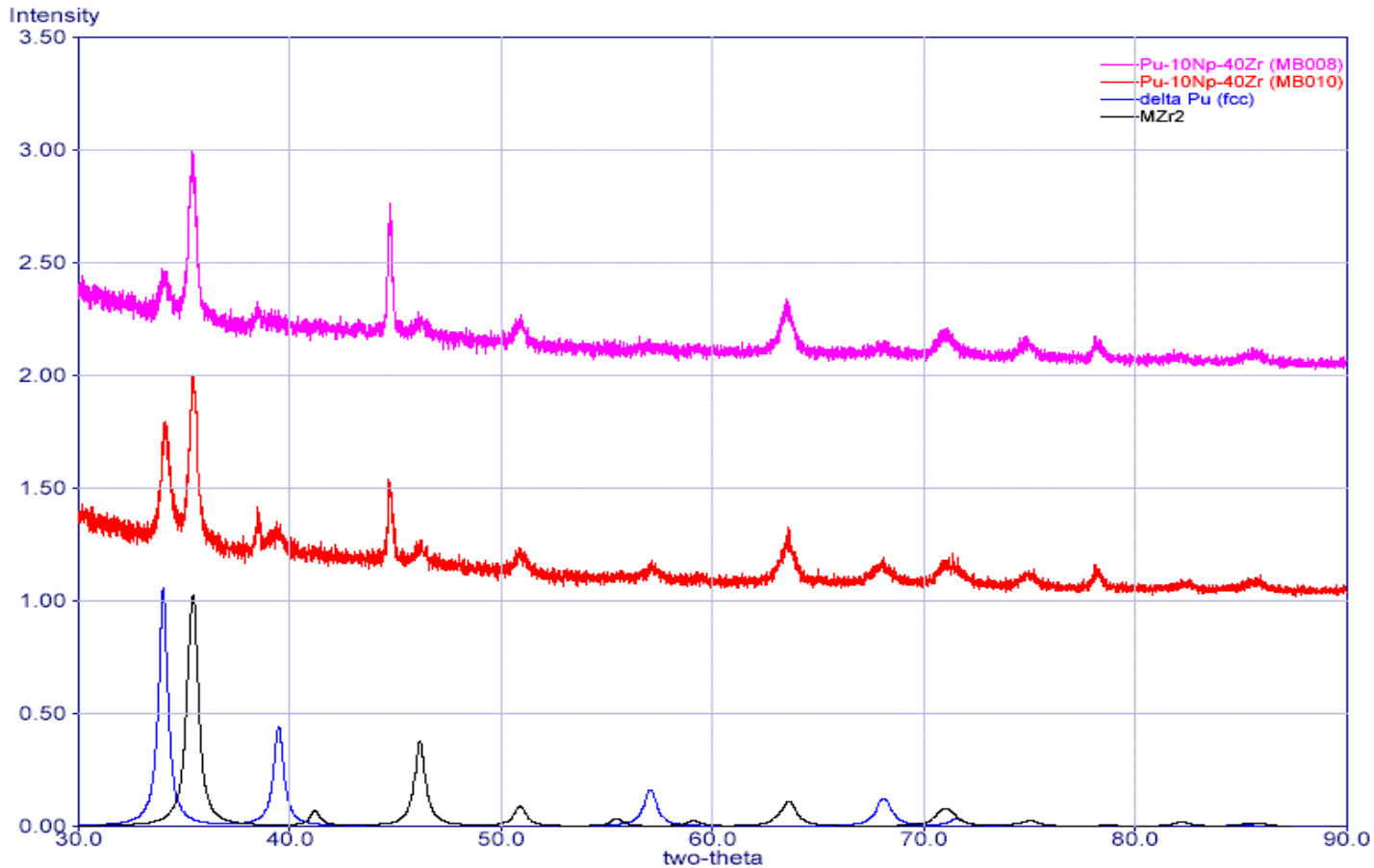
Backscatter Image



Oxygen



Powder XRD for Pu-10Np-40Zr



δ-$(Pu_{1-x}Np_x)Zr$: $a = 4.565$Å

δ-$(Pu_{1-x}Np_x)Zr_2$: $a = 5.055$Å, $c = 3.123$Å

Thermal analysis

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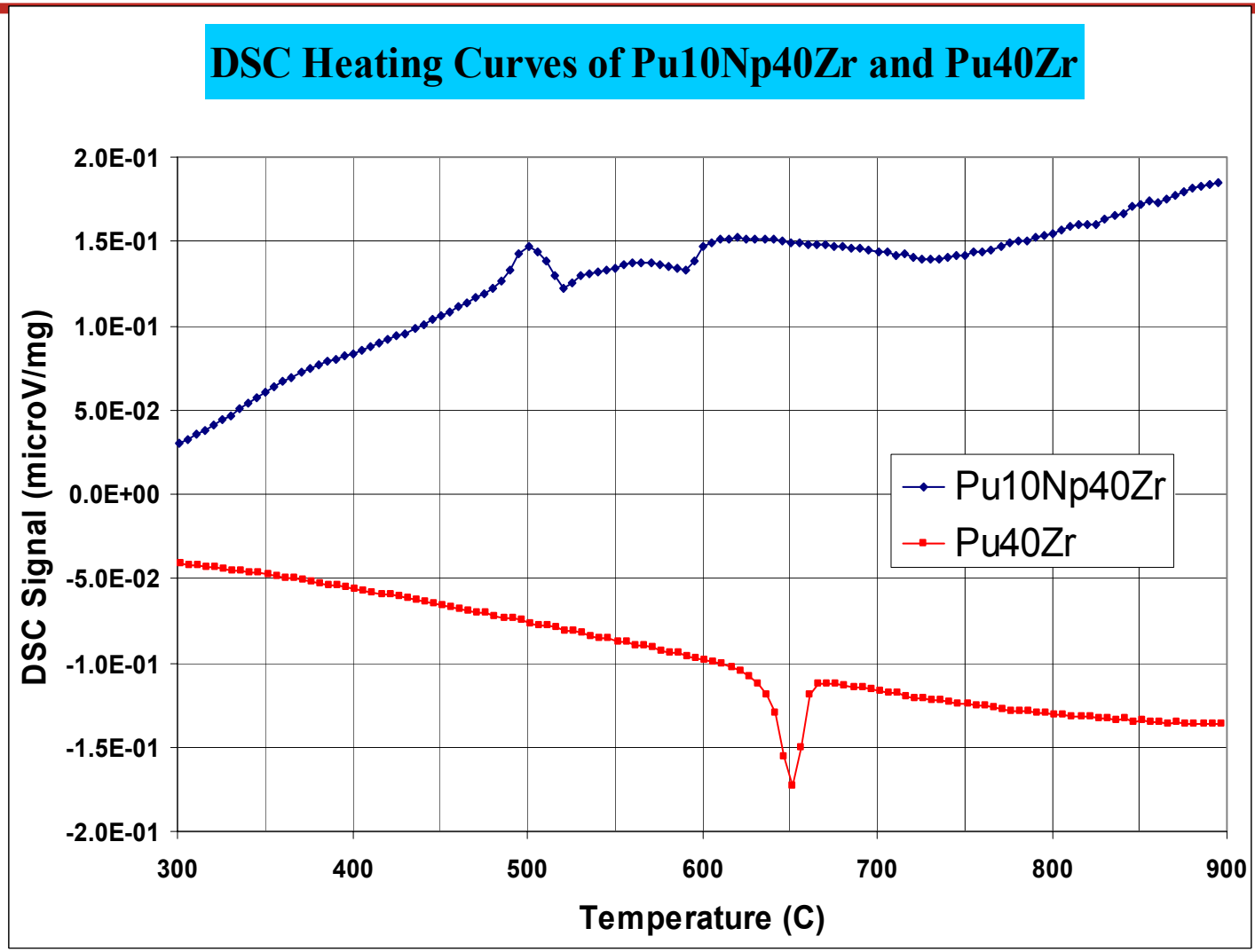
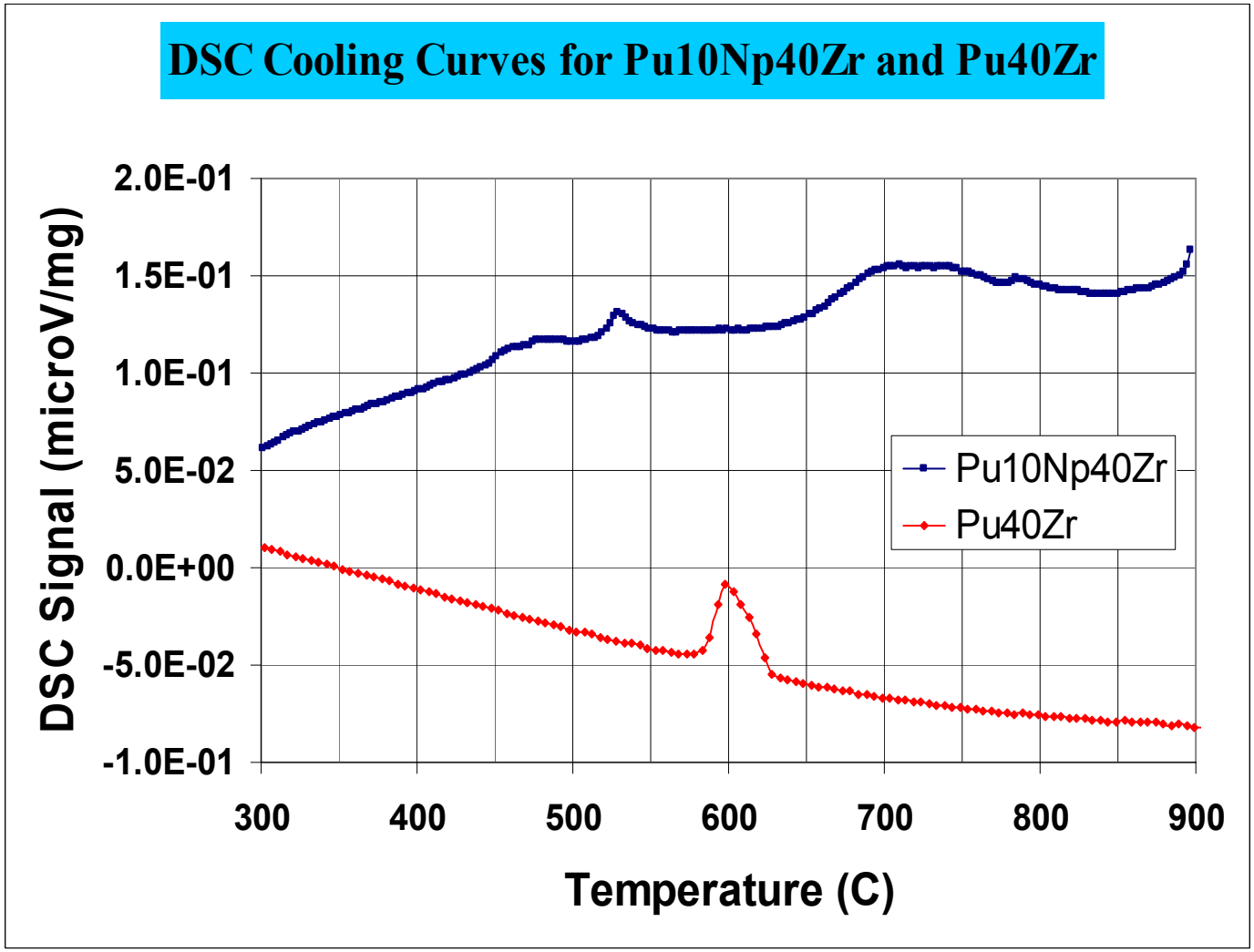
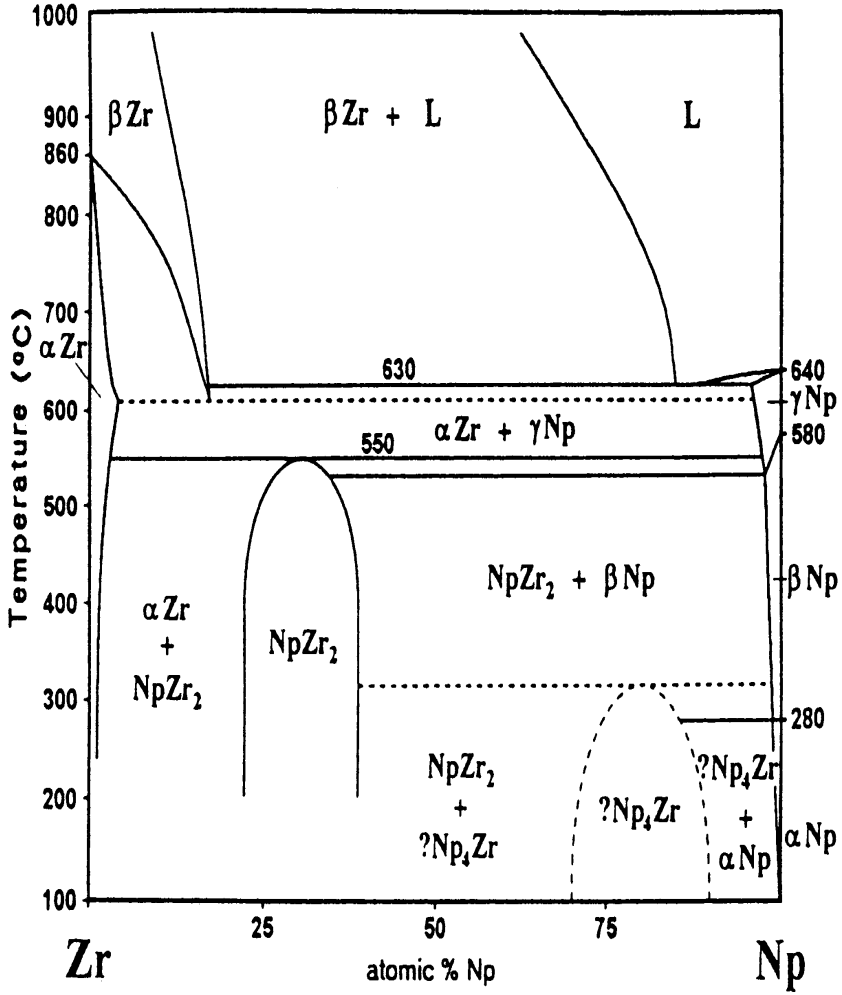
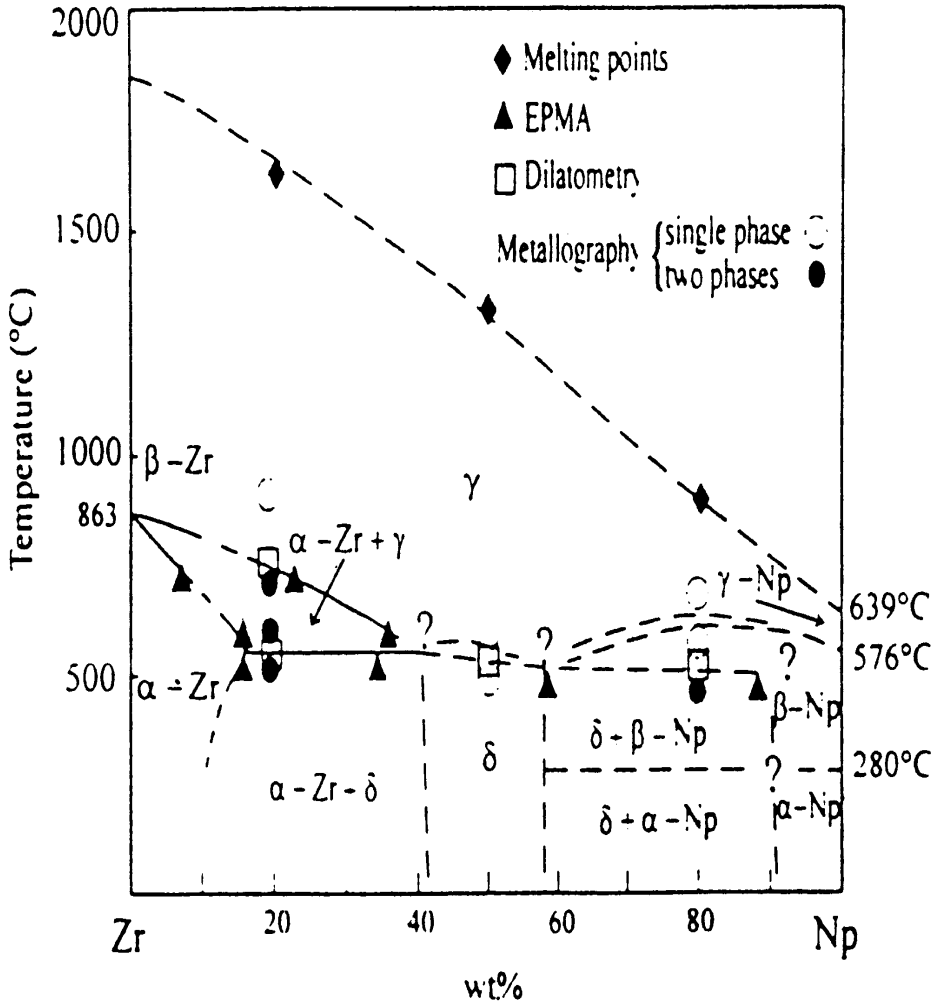


Thermal analysis



Proposed Np-Zr Phase Diagrams





AFC-1 Test Series Schedule

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- **Current AFC-1 Test Series Schedule**
  - **Likely delay of A-D due to basket and INEEL funding**
  - **Likely delay of E-H due to INEEL funding**
  - **ATR CIC scheduled for Feb-04, but slight delay is possible**

Test	Fuel Types	IRRADIATION TEST DURATION													
		2002	2003												2004
		DEC	JAN	FEB	MAR	APR	MAY	JUN	JUL	AUG	SEP	OCT	NOV	DEC	JAN
AFC-1A	Non-Fertile Nitride														
AFC-1B	Non-Fertile Metal														
AFC-1C	Non-Fertile Nitride														
AFC-1D	Non-Fertile Metal														
AFC-1E	Low-Fertile Nitride														
AFC-1F	Low-Fertile Metal														
AFC-1G	Low-Fertile Nitride														
AFC-1H	Low-Fertile Metal														

AFC-1A, -1B, -1C and -1D Fuel Test Matrix

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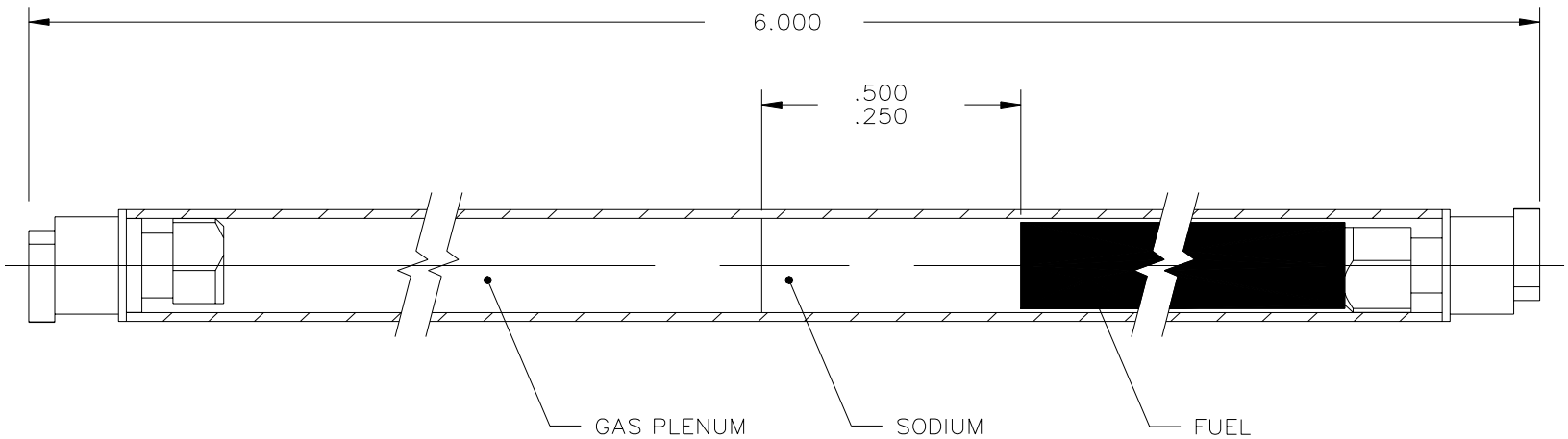
- **Six vertically-stacked rodlets per irradiation vehicle**
- **Low smear density, Na-bonded fuels to allow for swelling**

	EXPERIMENT	
Rodlet	ATW-1A & -1C	ATW-1B & -1D
1	$(Pu_{0.2}, Am_{0.8})N$-36ZrN	Pu-12Am-40Zr
2	$(Pu_{0.8}, Am_{0.2})N$-36ZrN	Pu-10Am-10Np-40Zr
3	$(Pu_{0.5}, Np_{0.5})N$-36ZrN	Pu-40Zr
4	PuN-36ZrN	Pu-12Am-40Zr
5	$(Pu_{0.50}, Am_{0.25}, Np_{0.25})N$-36ZrN	Pu-10Np-40Zr
6	$(Pu_{0.5}, Am_{0.5})N$-36ZrN	Pu-60Zr

AFC-1 Rodlet Design



AFC-1 Capsule Design

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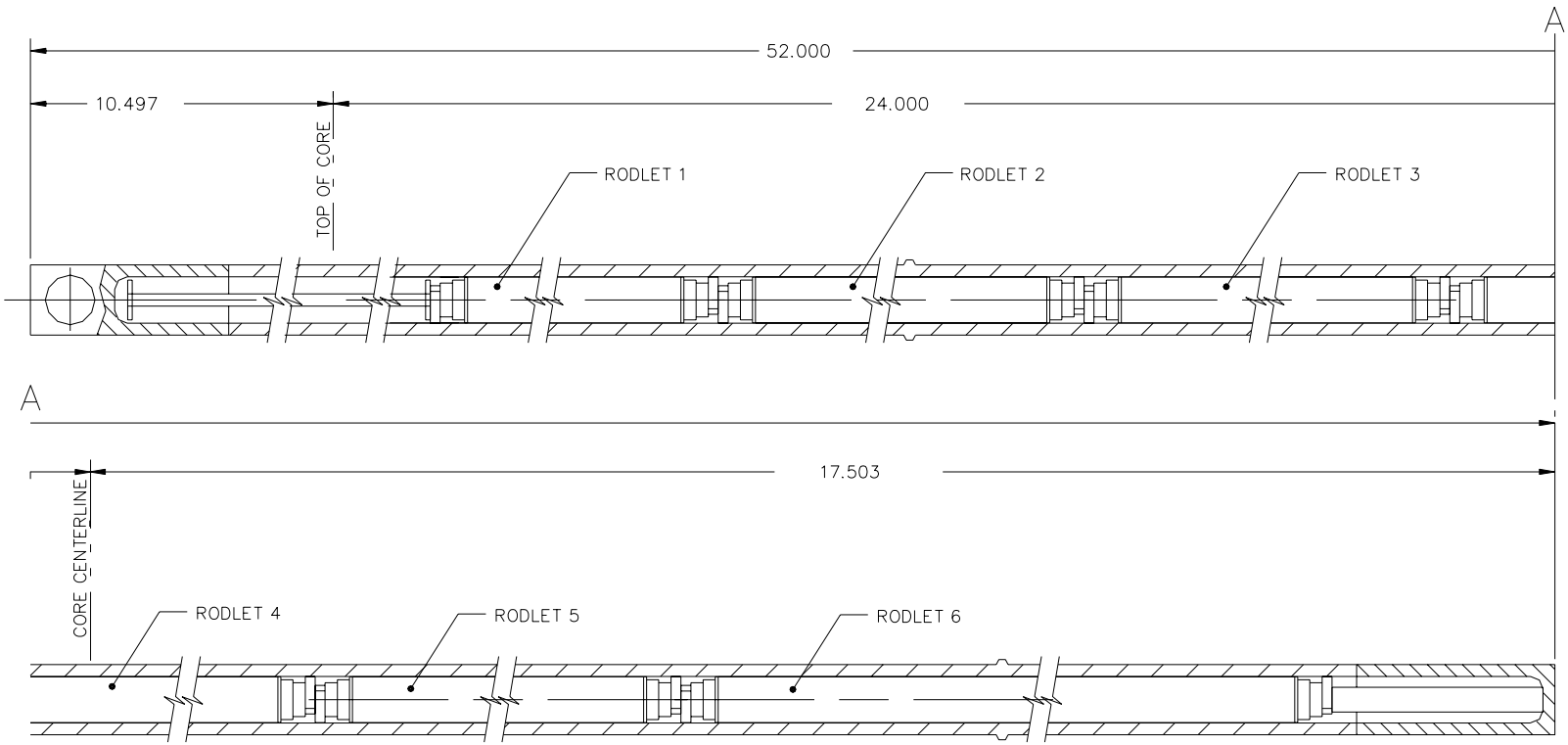

FUTURIX Irradiation Experiment

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- **International collaboration between DOE and CEA**
  - Collaboration in final stages of being formalized
  - Irradiation scheduled to begin in April 2006
  - Irradiation for 2 cycles (240 EFPD's) prior to Phénix termination
  - Discharge burnups ~8%; Am-241 transmutation >20%
- **Experiment design**
  - Will employ standard Phénix stainless steel cladding
  - Experimental fuel pin design (10 cm fuel column; large gas plena)
  - 8 test pins incorporated into 19-pin experimental subassembly
  - Fuels fabricated at ANL, LANL, CEA and ITU
  - Fuels encapsulated into pins at ITU
  - Experimental subassembly assembled at CEA

FUTURIX Irradiation Experiment (2)

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- **FUTURIX fuel test matrix**
  - Test revised to include non-fertile and low-fertile compositions
  - 2 metallic alloy fuels (Na-bonded) - ANL to fabricate
  - 2 nitride fuels (Na-bonded) - LANL to fabricate
  - 2 non-fertile oxide fuels (He-bonded) - CEA to fabricate
  - 1 fertile oxide; 1 cermet fuel - ITU to fabricate
- **FY-03 activities**
  - Finalize experiment design; submit Presentation Report
  - Fabrication facility preparation
  - Begin R&D phase on test fuels
    - » Establish test fuel fabrication process
    - » Characterize test fuel compositions
    - » QA Plan to be developed and approved

FUTURIX Irradiation Experiment (3)

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- **Future U.S. Deliverables & Milestones**
  - Input to Technical Report due April-04
  - Report on R&D Phase due Jun-04
  - Fuel Specification/Fabrication Control Plan due Sept-04
  - Fuel Fabrication complete Jun-05; Fabrication Report due Jun-05
  - Fuel Delivery to ITU in Jul-05
  - Input to Safety Report due Aug-05
- **Irradiation Begins April 2006**
- **Irradiation Ends February 2008**
- **PIE Complete with Final Data Exchange by October 2010**

Summary

- **AFC-1**
  - **fabrication mostly on schedule, with some minor issues to resolve**
  - **Basket problem being addressed by INEEL**
  - **CR funding for INEEL is an issue that can impact schedule**
- **Futurix**
  - **Schedule for preparation of experiment discussed in detail at DOE/CEA mtg on Jan 14, 15 and agreed upon**
  - **Some urgency is nearing for contract/agreement**
- **Series of Pu-Zr based alloys fabricated for AFC-1 irradiation test**
  - **Alloys have been encapsulated with Na in cladding tubes**
  - **Awaiting irradiation beginning in March 2003**
- **Powder processing did not produce desired product**
- **Arc casting worked well**
  - **Low americium loss due to short melt times**
  - **Good alloy homogeneity**
  - **Other processes possible for large-scale fabrication**
  - **Americium loss not an issue**

Summary (cont.)

- **Alloy characterization (results to date)**
  - **Pu-60Zr: α-Zr by XRD**
  - **Pu-40Zr: single phase δ-Pu**
  - **Pu-12Am-40Zr**
    - » **First data on this ternary alloy**
    - » **Single-phase δ-Pu**
  - **Pu-10Np-10Zr**
    - » **First data on this ternary alloy**
    - » **Two-phase fcc δ-(Pu,Np)Zr and hexagonal δ-(Pu,Np)Zr_2**
    - » **Oxygen impurities appear to have influenced microstructure**
    - » **Initial thermal analysis results are inconclusive regarding formation of low-melting-temperature phases**
    - » **Continued characterization to include TMA/dilatometry and high-temp x-ray diffraction studies**
    - » **Proceeding to obtain quality Np feedstock**

Review and Conclusions

- **Modified arc-melting technique acceptable for fabrication of experimental fuel pins and characterization samples.**
- **As-cast products in the "Pu-10Np-40Zr" regime identified as the fcc δ-$(Pu_{1-x}Np_x)Zr$ solid solution and the hexagonal δ-$(Pu_{1-x}Np_x)Zr_2$ phase domain. Pu-40Zr only fcc δ-PuZr.**
- **Oxygen impurities in feedstock charge may have and are suggested to have influenced products formed.**
- **Preliminary thermal analysis studies inconclusive as to melting behavior due to severe oxidation of sample.**
- **Continuing with thermal characterization to include thermo-mechanical/dilatometry and high temperature XRD studies.**
- **Proceeding to procure and/or purify quality Np feedstock.**


Vita
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